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**PAPER AND PRINTED PAPER SURFACE CHARACTERISTICS  
STUDIED USING AN OPTICAL METHOD**

**WANNA SANANPANICHKUL**

**A thesis submitted in partial fulfilment of the  
requirements of the Open University  
for the degree of Doctor of Philosophy**

**December 1993**

*LONDON INSTITUTE :*  
**The London College of Printing and Distributive Trades**  
**in collaboration with**  
**Coates Lorilleux International**

Author's number : PI 21667

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## **Associated studies.**

Additional studies of the subject area and related aspects include:

1) Attendance and participation in a number of relevant international conferences and Seminars.

- Pira Conference : Surface Treatment of Fibrous Webs, Lancashire, 18 - 19 April, 1989.
- Student Poster Paper Presentation entitled 'Printed (Lithographic) Paper Smoothness Studied by an Optical Method': at 1992 International Printing & Graphic Arts Conference, Pittsburgh, Pennsylvania, USA, 18 - 21 October, 1992.
- Tenth Fundamental Research Symposium of the Oxford&Cambridge Series (Products of Paper Making), Oxford University, England, 20 - 24 September, 1993.

2) Attendance at the meetings of

- The 3-P group of the Institute of Physics.
- The Royal Society of Chemistry.
- Pira International

3) Visits to various related exhibitions.

- Drupa 90 (International Printing and Paper Exhibition), Dusseldorf, Germany, 27 April - 10 May, 1990.
- IPEX 93 (International Printing Exhibition), Birmingham, England, 7 - 15 September, 1993.

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## **Units and Abbreviations.**

The units of this work are based on the SI convention. Abbreviations are those in common use. Those that require explanation are covered in text.

### terms.

rpm	revolution per minute
$\bar{X}$	arithmetic mean
S, S.D.	standard deviation
V	variance
C	covariance
C.I.	confidence interval
CV	coefficient of variation
RH	relative humidity
Min.	minimum
Max.	maximum

### quantities.

g	gramme
g/m <sup>2</sup>	gramme per square metre
m/min	metre per minute
ml/min	millilitre per minute
nm	nanometre
$\mu$ , $\mu$ m	micron
°C	degree celcius



# **PAPER AND PRINTED PAPER SURFACE CHARACTERISTICS STUDIED USING AN OPTICAL METHOD.**

**W SANANPANICHKUL**

A non-contact optical method for evaluating surface characteristics is reviewed. The optical reflectance instrument has been improved to be able to evaluate printed surfaces. Experiments were conducted with solid prints prepared on two types of papers printed with a heatset yellow ink. Both paper surfaces and printed surfaces are characterized into two regions: above the surface plane resulting in macrosmoothness ( $S_m$ ) and below the surface plane resulting in microsmoothness ( $S_u$ ). For a better understanding of such optical print smoothness, a printed surface model is proposed based on Barkas' classical model. It is generally known that the qualities of a print are determined by the materials and their interactions in the process, therefore the formation of printed surface characteristics has been discussed in relation to ink and paper interaction. Print smoothness is influenced by the uncompressed paper roughness and porosity, which determine the degree of ink penetration and ink distribution on the surface.

Offset lithographic printing has been the most widely used printing process, printing onto paper substrates. To achieve good press performance and high quality prints, the ink has to emulsify a certain amount of fountain solution; maintenance of this ink and water balance is, therefore, very important. The effects of fountain solution emulsified in the ink on print smoothness were investigated. A range, of varying amounts of fountain solutions, was emulsified in a heatset yellow ink using a high speed laboratory mixer; these 'emulsion inks' were printed as soon as possible after preparation. It was found that the print smoothness in macro regions, for both uncoated and coated papers, decreased significantly. In addition, an unpigmented ink system was employed to verify the role and the effects of pigment in the emulsification mechanism on print smoothness. The results indicated that pigment is the dominant contributor, to a smoothness decrease; and the pigment effect arises from the amount of fountain solution emulsified in the ink.

It has become important to measure print quality directly and quantitatively in the developments of ink, paper and printing technologies. This method makes it possible to measure print smoothness as a criterion for print quality.

## **AIMS OF THIS STUDY.**

Conventional measurements of print quality have been made on print density or print gloss. Few studies have been concerned with the characteristics of printed surfaces. This study concerns a measure of print quality in terms of print smoothness by an optical method. The aims of the study are

1. To improve an instrument so as to be able to measure the reflection of printed surfaces.
2. To verify the method for reliable determinations of print smoothness. This requires a large number of samples to be examined and statistical methods employed to ensure significant data. When this has been achieved, a printed surface model can be proposed.
3. To investigate the effects on print smoothness, of materials involved in the offset lithographic process, on a laboratory scale printing press.

Print smoothness is one of the most important print quality factors. It is hoped that this study will provide a better understanding of ink / paper interactions; and, for the offset lithographic process, 'emulsion ink' / paper interactions. The method of this study may be useful to both ink makers for the developments of their ink formulations and printers to be aware of such printed defects which may occur.

## 1. INTRODUCTION.

The products from any printing process are prints whose properties and characteristics correspond to the desired results. A study of ink and paper interactions is obviously important since they determine not only the efficiency and quality of a printing process but also the qualities of the prints. Ink and paper interactions are two-phase interactions involving ink transfer in the printing nip and ink drying after the nip. Various phenomena such as contact between ink and paper, ink immobilization, wetting and spreading of the ink, ink film splitting, ink penetration, determine the characteristics of a printed surface. DeGrâce & Dalphond [1989] stated that the smoothness and the uniformity of the prints are determined by the uniformity of pigment distribution on the surface, the penetration and the evaporation of ink vehicles and the film formation of the polymer which holds the pigments within the ink film.

Earlier researches on print smoothness have been done through the use of specular gloss measurements or goniophotometer measurements. For example, Leekley, Denzer & Tyler [1970] employed a goniophotometer from which they could separately measure specular and diffuse reflectance of paper and printed surfaces. Their optical smoothness results were shown by the distributions of the inclination angles of surface elements relative to the nominal plane of the samples. They suggested that the change in distribution of inclinations between unprinted and printed samples provided information that paper surface irregularities were filled with the inks.

In her study of surface reflection of coated papers and prints, Oittinen [1980] investigated the effects of ink variables on ink transfer and print gloss. Print gloss measurements were determined using a gloss meter which is in accordance with the TAPPI standard method. The influences of ink film thickness on paper smoothness and print gloss were explained through filtration and absorption mechanisms.

As demonstrated by Maley [1990], ink film smoothness and ink uniformity are the major contributors to print quality. However, there are not many methods available that are capable of providing good information on printed surfaces. One reason is that printed surfaces are the products of many factors in the printing process. The present study is concerned with the printed surface characteristics of solid prints in terms of print smoothness by using the non-contact optical method, which was previously employed to evaluate paper surface smoothness [Hansuebsai, 1989]. The sensitivity of that method was increased, so as to provide valid results by the improvement of its instrument and by an appropriate increase in the number of samples evaluated. A printed surface model based on Barkas' classical model is proposed as an ink vehicle-air interface rather than a pigment-air interface. The printed surfaces are characterized into two regions. One region is at or above the surface plane corresponding to dried polymer film, which results in macrosmoothness ( $S_m$ ). The other is below the surface plane corresponding to the dried polymer film and the pigments within the ink film, which results in microsmoothness ( $S_u$ ).

The offset lithographic process is the most widely used printing process and dominates the printing of papers and boards. The uniqueness of

the process is that the image and the non-image areas are in the same plane. During printing, fountain solution is applied to non-image areas to keep these areas free from inks whereas ink is applied to image areas. As a consequence, the quality of the printed image produced by the offset lithographic process is determined by the combination of three major materials and their interactions: ink, paper and fountain solution. It is well known that offset litho inks emulsify a certain amount of fountain solution during printing and thus the maintenance of ink and water balance is the most crucial requirement to achieve high print quality and to avoid printing problems. Most of the printing problems are related to too much water. Extensive studies [e.g., Störm & Vanderhoff, 1984; Fetsko, 1986; Surland, 1980, 1983] have been made on ink and fountain solution (water) interactions on the plate. Few works have been concerned with the effects of emulsification on the resultant prints. Therefore, this study was aimed to investigate the effects on print smoothness, of fountain solution emulsified in the ink. To simulate offset lithographic printing, a 'pre-emulsified ink' was prepared using a high speed laboratory mixer [Bassemir & Krishnan, 1991]. A heatset yellow ink was chosen to be studied and the amount of fountain solution emulsified in the ink has been varied. Printing of these 'emulsion inks' was carried out as soon as possible after ink preparation on a laboratory printability tester under controlled conditions. Statistical analysis of the results was employed to indicate the significance of the effects and the results were discussed in relation to 'emulsion ink' and paper interactions.

In addition, the role and the effects of pigments on the emulsification mechanism in this process are not fully understood; unpigmented ink, whose formulation is similar to the heatset yellow ink with the pigment excluded,

was also prepared to study the effects on print smoothness. The emulsification, water uptake tests, for both heatset yellow ink and unpigmented ink were carried out using a modified Surland's method to provide supportive information.

Surfaces of all unprinted and printed samples were also examined by a scanning electron microscope. The results not only provided a visual image of each surface but also provided supporting evidence for the effects on print smoothness.

## **2. BACKGROUND.**

### **2.1 BACKGROUND ON OFFSET LITHOGRAPHY.**

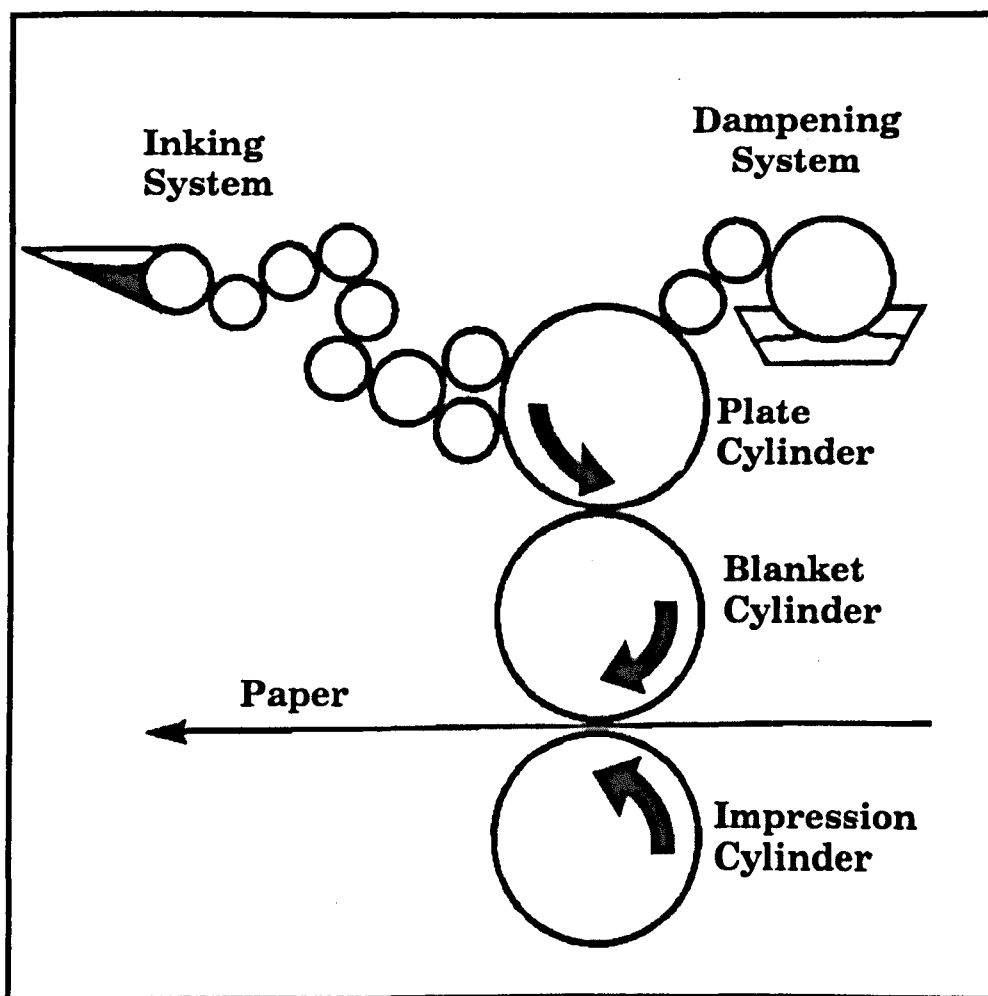
Printing is the process of making copies of graphic images and text that are usually transferred from an image carrier or printing plate to a substrate such as paper using a printing press. Among the major printing processes, offset lithography is the most widely used process and its major application is for printing on paper and board substrates. The name 'offset' is given because the inked image does not transfer directly from the image carrier or plate to the substrate, but it is 'offset' from the plate onto a rubber blanket and then to the papers or other substrates.

For a better understanding of the process, it is necessary to investigate the operation of the printing press. The major components of an offset lithographic press are the feeder unit, the printing unit and the delivery unit. The most important component is the printing unit. Figure 2.1 shows a diagram of a typical offset lithographic printing unit.

Every offset printing unit consists of a plate, blanket and impression cylinder together with dampening and inking systems. Since it is the area where ink and paper interactions occur, some of these will be discussed in detail.

#### **2.1.1 The inking system.**

This is generally made up of two types of rollers, namely rubber covered and hard surfaced rollers. A large number of rollers are arranged



**Figure 2.1 A diagram of a typical offset lithographic printing unit.**



alternately in a train to break down the ink from the ink reservoir into a thin film on the image area of the plate. Ink film thickness needs to be accurately controlled since a variation as little as 0.1 micron in a one micron film thickness on paper can cause variations in print density of  $\pm 0.1$  [Martin & Silver, 1976]. MacPhee [1979] proposed an idealized model for an inking system using a roller train assuming a 50/50 film splitting at any ink roller nip. He showed that the ink film thickness of approximately 2-3 microns and the fountain solution film of approximately 0.5-1 microns carried by the plate are necessary to obtain an ink film of 1 micron on the print.

### **2.1.2 The dampening system.**

The function of the dampening system is to provide and maintain a film of fountain solution on the non-image area of the plate. Studying surface energy, phase interaction and work of adhesion of two lithographic plates, Ström & Vanderhoff [1984] concluded that during printing the non-image area carries a layer of bound fountain solution and a layer of free fountain solution. Both the fountain solution film thickness and the ink film thickness are very important to the success of the offset printing process. Scumming results from too little fountain solution on the non-image area whereas water marking results from too much water on the ink layer of the image area. It has been reported that the fountain solution film thickness on the plate is about one micron [Lawson & Watkinson, 1975]. As previously described, MacPhee [1979] considered that this fountain solution thickness is approximately one-half or one-third of the ink film thickness on the image area of the plate. This proportion is normally referred to as the appropriate ink and water balance and is dependent on the ratio of image to non-image areas.

### 2.1.3 Plates.

The uniqueness of offset lithography is that it is a planographic process, i.e., printing is achieved from a flat or plane surface. This can be achieved based on the fact that oil and water are immiscible. According to this, the basic requirement of the plate is an ability to produce an image area to accept only ink and repel water (hydrophobic) and a non-image area to accept water (hydrophilic). Various methods are employed to produce plates with this requirement, i.e., to produce a printing plate with completely different surface energetic properties between the image and non-image areas.

To understand the process of producing an inked image on the plate it is necessary to study surface tensions and contact angles of inks, fountain solutions and plates. Surface tension and contact angle are discussed adequately in many textbooks and so only a brief discussion is given here.

Molecules in any materials attract each other. The molecule within the bulk of the material will be attracted in all directions by its neighbours, therefore there is no tendency to move in any one direction. A similar molecule at the surface has only half the number of neighbours attracting it. Thus, there is a force pulling it, and all other surface molecules, inwards which can contract the surface area. The potential energy produced in all the surface molecules constitutes surface energy and the force trying to contract the surface area is the surface tension. The two are virtually equivalent. Surface energy and surface tension exist not only at the surface of a liquid and air, but also at the surface of solids, at the interfaces between a liquid and a solid and *vice versa*. The units for surface tension and surface energy

are different but equivalent. For this thesis, surface energy will generally be used in reference to solid surfaces and surface tension for liquid surfaces.

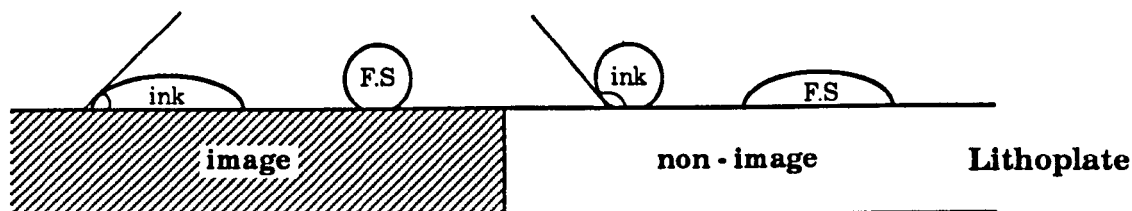
When a drop of liquid is placed on a solid surface, it may either spread across the surface or may remain as a drop having a finite contact angle ( $\theta$ ). The contact angle is dependent on the surface energy, the phases interact according to the Young-Dupre' relation.

$$\gamma_s = \gamma_{sl} + \gamma_l \cos \theta \quad \dots\dots\dots(1)$$

where

- $\gamma_s$  is the solid surface energy
- $\gamma_l$  is the liquid surface tension
- $\gamma_{sl}$  is the solid/liquid interfacial energy

If a liquid with a higher surface tension is placed on a solid surface with a lower surface energy, the liquid will form into globules and will not wet the solid surface. In terms of contact angle, it will show a large contact angle. On the other hand, if the liquid has a lower surface tension than the solid surface, the liquid will wet and spread across that solid surface. The contact angle will be low and approaching zero. On a printing plate, the image area should show contact angles approaching zero for the inks whilst the non-image area should show contact angles approaching 180 degree for the inks (Figure 2.2).



**Figure 2.2 Lithoplate Surface Characteristics.**

Most solid surfaces used to make offset plates can be wetted to some extent by the inks used. Therefore, almost all offset plates require an application of fountain solution to maintain the distinction between the image and non-image areas. The main component of a fountain solution is water which is a polar liquid having a high surface tension (ca 72.8 dynes/cm). In contrast, lithographic inks are virtually non-polar liquids having lower surface tensions approximately 30-35 dynes/cm [Banks, 1965; MacPhee, 1979]. On the press, the plate is clamped round the plate cylinder where it is first damped by the dampening form rollers. Because the surface tension of fountain solution is lower than the surface energy of the non-image area of the plate, fountain solution will form a thin film on the non-image area. On the other hand, it will contract into tiny droplets on the image area because of its higher surface tension compared to the surface energy of the image area of the plate. Ström & Vanderhoff [1984] found that after a film of fountain solution was applied, the wetted plate has the same surface energy as the fountain solution used.

When the wetted plate passes under the ink form rollers, these rollers cannot ink the areas already covered with a film of fountain solution, i.e., the non-image area. This is because hydrophilic substances such as gum arabic

are present not only on the non-image area of the plate but also in the fountain solution. This promotes the attraction of fountain solution to the non-image area. This area thereby retains its hydrophilic property so that the ink cannot displace the fountain solution and adhere to the non-image area. Although ink and fountain solution are immiscible, in the nip between the ink form rollers and the plate (Figure 2.1), an emulsification process occurs. Stefan's theory [1874] explains a simple model of a liquid confined by two flat parallel plates. The force required to separate the plates is expressed as follows: [MacPhee, 1979]

$$F = \frac{C\mu VA}{t^3} \dots\dots\dots(2)$$

- where F = force
- C = constant
- μ = viscosity
- V = speed at which the plates are separated
- A = plate area
- t = distance between the plates (i.e., film thickness)

This equation shows that the force required to split a liquid increases with increasing liquid viscosity. Tollenaar [1973] and Karttunen & Manninen [1978] stated that at the nip exit, film splitting occurs in the much lower viscosity fountain solution film rather than in the ink layer. Thus, a film of fountain solution will remain on the non-image area but some droplets may transfer to the ink which are subsequently emulsified with the ink. In other words, the ink does not form a film on top of the fountain solution film. This has been shown by Ström & Vanderhoff [1984] who found

that the work of adhesion of ink on the non-image area is close to zero and independent of the surface tension of fountain solution. On the other hand, the ink form rollers will ink the image area either by pushing aside or emulsifying the droplets of fountain solution to make an inked image.

The inked image is now transferred onto the rubber blanket, where the image is reversed to a mirror image. The substrate is then passed between the blanket and the impression cylinders where the image becomes right-reading again. The printing pressure is applied by the impression and the blanket cylinders in order to transfer the image from the blanket to the substrate. The correct pressure is of importance to print quality. Therefore, accurate adjustment of the gap between the blanket and the impression cylinder is necessary for each substrate. Details of the ink transfer to paper will be discussed in the section on ink and paper interactions.

## **2.2 BACKGROUND ON PAPER.**

Paper can be defined as a continuous web of materials formed by the deposition of vegetable fibres, synthetic fibres or a mixture of fibres with the addition of other substances, from liquid suspension so that the fibres are intermeshed and bonded together. Paper may be coated, impregnated, printed or otherwise converted, during or after its manufacture, without necessarily losing its identity as paper.

### **2.2.1 The manufacture of paper.**

Papermaking can be divided into 4 distinct stages: the manufacture of pulp, the preparation of stock for the papermachine, the papermaking operation and the paper after-treatments and finishing. The nature of the fibres used and the treatments at these 4 stages determine the properties of the finished paper, particularly the printing characteristics [Grant & Young, 1978].

*The manufacture of pulp.* This is known as 'Pulping process'. This stage is the separation of cellulose fibres from undesirable materials associated with them in the plants. Cellulose fibres from wood have become the most important source of pulp for papermaking. There are two basic groups of wood namely softwood and hardwood. In softwood, there is virtually one type of fibre whereas hardwood has a more complicated structure with both long, or short and thick fibres. The chemical composition as well as the dimensions of the fibres are different. The pulping process makes use of the differences in physical and chemical properties of cellulose and lignin to separate them. In the mechanical pulping process, the fibres are liberated from wood by physical degradation of the middle lamella

between the fibres. The resulting pulp is a mixture of bundles of fibres and non-fibrous materials that existed in the original wood. Chemical pulping process extracts lignin and other impurities from the wood by chemical solution. This process yields the cellulose fibres in a separated and relative pure state. Fibres extracted from hardwood are normally shorter than those from softwood.

*Stock preparation.* This is the most important stage in papermaking because the majority of the paper properties are determined by the stock preparation. This covers the processes of producing 'stock' that is, a suspension of fibres and other non-fibrous materials in water, in the appropriate condition and composition for the production of a desired paper. The most important process is beating or refining which is a mechanical treatment to modify the fibre properties. There are three most important properties which are normally modified by the beating or refining operation:

1. changes in fibre length as a result of cutting or shortening the fibres.
2. flexibility resulting from internal fibrillation that is bruising and splitting the fibres internally.
3. changes in specific surface defined as square metres of surface per gramme of fibre affected by external fibrillation.

The original physical and chemical properties of the fibres used, the equipment, the temperature and the consistency of the pulp, all influence the effects of beating or refining, which in turn determine the main characteristics of the paper. Papers made from unbeaten pulp show less tensile and burst strength; and are bulkier with high porosity, absorbency, opacity and uneven formation. On the other hand, papers made from beaten



pulp exhibit greater strength, a higher density, a smoother surface and a more uniform formation.

A number of non-fibrous materials are also added to the stock to obtain particular paper properties. The most important of these are sizing agents and fillers. The purpose of sizing is to improve the water resistance properties of paper. The most widely used method is internal or engine sizing. This is achieved by adding rosin and aluminium sulphate (alum) to the stock. Alum will precipitate particles of aluminium rosinate as a coating of size onto the fibre walls. This reduces the capillary action of water and prevents penetration into the paper.

Fillers or loadings are water-insoluble, white mineral pigments originally added to modify characteristics of the finished paper. By filling in the interstices of the fibre network, they give a smoother surface. They also impart some specific properties such as brightness, opacity, softness and dimensional stability to the paper. However, due to their interference with fibre bonding, they appear to reduce the strength and degree of sizing.

*The papermaking operation.* Formation of the paper web takes place in the papermachine. The main function of the papermachine is to remove the water from the stock so that the fibres come together to form a web of paper. In the papermachine the stock is passed from the headbox through the slice onto the wire where the water is removed from the stock. The consistency or concentration of fibres of the stock affects sheet formation. A lower consistency tends to improve sheet formation [Hallgren & Lindström, 1989].

Formation is the term used to describe the distribution of fibres within the finished sheet. Bad formation results in uneven ink absorption when printed.

Water removal in the papermachine is accomplished by free drainage, by suction on the wire section, by pressing on the press section and by evaporation in the dry section. Throughout this sequence of water removal, different phases of consolidation of the web occur [Rance, 1980]. From the wire section to the press section, there is formation and compaction of the suspension. Most of the water is taken out on the wire section, hence it is on the wire section where the web of paper is first consolidated and many of its characteristics established. In addition, some of the characteristics of the paper are formed on the press section as a result of further consolidation. This depends on the type of press and the nip pressures. As the paper web passes through the dryer section, it tends to shrink as a whole. The extent of shrinkage depends on the basis weight. Heavier webs tend to shrink more than lighter webs, resulting in different surface structures [Corte & Herdman, 1975].

*Paper after-treatments and finishing.* The term 'finish' describes the final surface or texture of a paper. Calendering in the papermachine is the last stage of consolidation, compressing the web which has already become a solid. It is considered to be the fine adjustment of the final characteristics of the sheet. The main purpose is to produce an evenness of surface as required by subsequent printing or coating processes. The surface of an uncalendered sheet is too rough to facilitate high quality printing. Calendering also improves the imperfections from the earlier stages of formation and consolidation to give uniformity of web thickness. The other purpose is to

impart specific optical properties to the surface of the paper, such as gloss. In some cases, smoothness and gloss are accomplished through supercalendering which is normally carried out as a separate process off-machine.

### **2.2.2 The coating of paper.**

The above description is how uncoated papers are generally manufactured. High quality printing requires a surface that is smoother than that resulting from papermachine alone. Therefore the sheets are often coated with a thin layer of mixture to fill in the voids and to cover the fibres. This helps to improve such properties as smoothness, gloss and other printing properties including ink absorptivity and ink holdout. Although a layer of coating can hide to some extent the irregularities of the base paper, the base paper must be uniform in its substance and texture, otherwise, the coating will mirror the surface contour. Coated papers generally have a denser and more uniform structure than uncoated papers. Most of the coated papers are calendered or supercalendered after the coating is dried. The effectiveness of the coating process depends on the formulation of the coating mixture, the amount applied onto the paper, the application method and the amount of calendering or supercalendering. Normally, matt-coated papers have little or no calendering, dull-coated papers are moderately calendered and gloss-coated papers are heavily calendered.

## **2.3 BACKGROUND ON INK.**

Printing inks are coloured materials applied to any substrate by one of the printing processes. They are therefore classified by the printing process used: letterpress, offset lithographic, flexographic, gravure, screen or other. It is the offset inks that are of interest in this study.

### **2.3.1 Ink composition.**

In common with other inks, offset inks are composed of three major components: pigments, vehicle and additives. Variations in these three basic components lead to differences in ink properties.

*Pigments.* These are finely divided solid colouring materials which are dispersed in the ink vehicle. Pigments impart colour or tone, opacity or transparency, and other properties to an ink. The nature of offset lithography imposes restrictions on the pigments that can be used. They must have a greater affinity for oil than for water, i.e., they must be insoluble in and unreactive with the fountain solution used. Offset lithography prints thinner ink films than any of the other major process mainly because of the low capacity of the plate for carrying ink. It follows that for offset inks to print a full colour strength, they must be formulated with high pigmentation. In general, the properties required for an offset ink pigment are good colour strength, reasonable stability to chemicals and fine particle size; with the ability to be dispersed in various types of offset ink vehicles and to give inks the desired rheological properties.

*Vehicles.* The main function of ink vehicles is to provide a means of transferring the pigments to the substrate and binding the pigment layer on

the surface of the substrate. The vehicles should not react with the pigments but should disperse them effectively and yield inks of good rheological properties. Additionally, they should be capable of emulsifying water or fountain solution but not to an excessive extent. They are made up of resin, oil and solvent of which the proportions are determined by the printing process and the drying system. Offset ink vehicles can be classified into two groups; oleoresinous and acrylate systems. The oleoresinous system, normally made up of hard resin and a drying oil alkyd, is used in producing quickset, heatset and oxidative drying inks. The acrylate system is used in radiation curing inks [Leach, 1988].

1. Oils. They can be classified into three groups; drying oils, semi-drying oils and non-drying oils. Usually, they are treated, purified or modified with other materials and used for different functions in ink formulations. Some are used as the raw materials in the manufacture of resins, or as solvents or diluents in the production of ink vehicles. Others are used as plasticisers or lubricants in inks.

2. Resins. These are non-crystalline solid materials or high molecular weight liquids with the ability to be dissolved in organic solvents. They impart hardness, gloss, adhesion and flexibility to an ink. The choice of the resin used has to meet fundamental requirements such as solubility in weak solvents and controlled water tolerance, i.e., neither fully water miscible nor totally repellent. They also need to be cohesive when used in a suitable solvent system.

3. Solvents. These are liquids capable of dissolving resins or oils. The usefulness of a solvent depends on its solvent power, that is, its effectiveness in dissolving polymers such as resins, oils or waxes. Another important property is the rate of evaporation because the solvents are normally required to leave the ink film immediately after printing. Offset inks also have to use solvents of low volatility to obtain press stability. The most important solvent is high boiling petroleum distillate. The two major properties of distillates are their boiling range and aromatic content. The boiling range affects press stability; a narrow range gives optimum press stability whilst a wide range leads to poor press stability. The aromatic content determines its solvent power that is the solvents having high aromatic content will have high solvent power.

*Additives.* A number of additives are added to impart special characteristics to an ink. It is essential that the additives are compatible with the ink. Plasticisers are added to make the resins more flexible and to promote binding of the ink to the substrate. Waxes are incorporated into the ink to impart the dried ink film with more slip, scratch and rub resistance. Driers are used as catalysts to promote drying by oxidation/polymerization of inks containing drying oil derivatives.

The formulation of a printing ink must meet a number of criteria. Offset inks have to be formulated to run in the presence of water or fountain solution. This necessitates the formulation of offset inks that take up a limited amount of water or fountain solution only as a finely divided dispersion. They should not tend to form an oil-in-water emulsion. In addition, offset printing machines are constructed in two basic types;

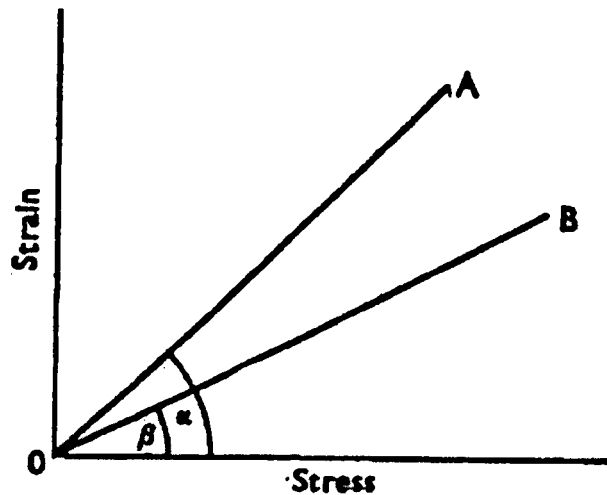
sheetfed and webfed presses. The printing speed of a webfed press is faster than that of a sheetfed press. This makes the formulations of offset ink more complicated.

The objective in ink manufacture is to produce a thorough dispersion of pigment particles in a varnish. Manufacture of offset inks usually takes place in two stages: mixing and milling. Mixing is a process in which ink components are mechanically mixed so that the pigment is initially wet and the aggregates of pigments are reduced in size. Milling is a further process to bring the ink to a smooth and homogeneous state ready for use in a printing machine [NAPIM, 1980].

### **2.3.2 Ink properties.**

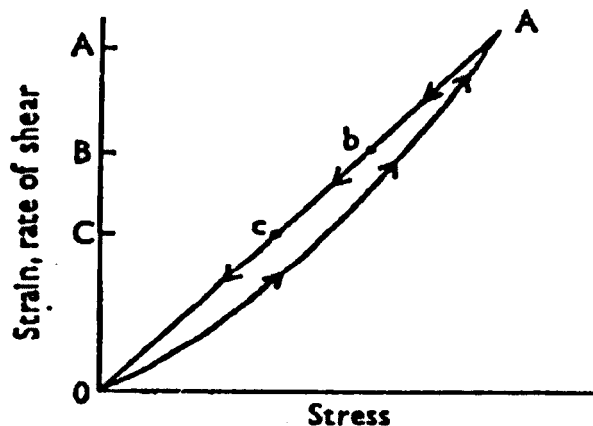
The ink flow properties are of great importance in offset lithographic printing. For an ink to transfer from one surface to another, until its transfer to paper, ink films are required to split. The properties of the ink which involve ink flow and ink splitting are viscosity and tack.

Viscosity is a liquid's resistance to flow. It is defined as the ratio of shear stress (shear force per unit area) to shear rate (velocity gradient of flow). Liquids such as the solvents used in the inks or water in fountain solution are examples of Newtonian liquids. Newtonian behaviour is where the flow is proportional to force applied and even a small force produces some flow as shown in Figure 2.3.



**Figure 2.3 Rheogram for Newtonian liquids.**

When pigments are mixed into Newtonian liquids, the resultant mixture has properties that differ considerably from Newtonian behaviour. Offset inks have a viscosity which is broken down by shearing but which rebuilds itself when the ink is allowed to stand. This is known as thixotropy which is a time dependent phenomenon. The diagram of the viscosity for a thixotropic system is shown in Figure 2.4.



**Figure 2.4 Viscosity for a thixotropic material.**



Offset inks are visco-elastic materials that is they contain both the viscous characteristic of a liquid and the elastic characteristic of a solid. The viscous liquid moves when a minimal stress is applied and the change persists on removal of the stress, whereas the elastic solid will recover its original form on removal of the stress.

Tack is defined as the force required to split an ink film. Tack is a critical factor influencing ink transfer to the paper. The tack of an ink must be lower than the surface strength of the paper so that the ink will not pick the paper at the required printing speed. In halftone printing, the tack should be at an optimum value. An ink having too high a tack may produce sharper dots or the actual printed images are smaller than the plate images resulting in loss of detail and colour balance. On the other hand, an ink having too low a tack may cause dot gain. Tack is more important for multicolour printing because the degree of ink trapping will depend on the relative tacks of the inks. In order to obtain a satisfactory printing, the tack of the first ink must be greater than that of the second ink which in turn must be greater than the tack of the third ink and so on.

### **2.3.3 Ink drying mechanisms.**

Printing inks are usually applied in the liquid phase to a substrate. Ideally these liquid inks should be stable both in the tin and on the press; but, as soon as possible after impression, should undergo a change from a liquid to a solid dried film. This change of state is referred to as ink drying and involves the formation of a film. Drying occurs by either a chemical or physical change in the ink or the combination of both physical and chemical changes [Askew, 1969].

### *Chemical film formation.*

1. Oxidation/polymerization. This is a chemical reaction in which many small molecules react to form large molecules or polymers. Oxidative/polymerization is a classical drying process for offset inks which involves oxygen-induced free radical polymerization. That is the atmospheric oxygen attacks at the activated sites on the drying oil chains in the resin system producing hydroperoxide which will decompose into free radicals. The free radicals of one molecule add on to another molecule of the drying oil increasing the molecular weight. There is also more than one activated site to be attacked, therefore cross-linkage occurs. These processes cause the ink vehicle to become a dried film. The dried ink film must have hardness, adhesion and flexibility during its lifetime. In summary, the process proceeds by four stages:

- a) Initiation or peroxide / hydroperoxide formation.
- b) Propagation or decomposition to form free radicals.
- c) Polymerization.
- d) Termination.

2. Radiation curing [Morrison, 1990]. This is an accelerated ink drying process involving an initial input of energy being transmitted from a suitable emitter and absorbed directly by the ink vehicle molecules within the wet ink film. The term 'cure' generally refers to the forming of chemical bonds in the drying of an ink. Free radical chain polymerization is enhanced by some forms of radiation such as ultraviolet, infrared, electron beam, microwave or radio frequency. The resins used contain highly reactive sites which create rapid chain reactions and thus a highly cross-linked dry film is produced in fractions of a second.

*Physical film formation.* The important physical processes are:

1. Absorption or penetration of the ink. This is the simplest physical method of drying. The ink does not really become solid in this case. The ink is absorbed into the network of narrow channels or pores of the paper by capillary action. Coldset inks based on hydrocarbon oils and resins are normally used in newsprinting. They dry solely by this mechanism and the drying speed is influenced mainly by the oils. Absorption drying is often used in conjunction with other drying processes.

2. Quickset mechanism. This drying process involves a combination of physical and chemical film formation namely absorption and oxidation/polymerization. Quickset ink vehicles consist of a viscous solution of hard resin in drying oil and very low viscosity petroleum distillate. Both components have to be sufficiently compatible to yield an ink with reasonable press stability. After the ink is applied on an absorbent paper, the capillary action mechanism draws the low viscosity distillate into the pores of the paper to separate it from the rest of the ink. The hard resin in the drying oil is therefore left on the surface. At this stage, the ink film is not fully dried. This is known as 'ink setting'. After the print has set oxidation/polymerization proceeds within the drying oils or the resins, leading to the formation of a three dimensional cross-linked network of chemical bonds in the ink film.

3. Evaporation. In this process volatile solvents or mixtures of solvents evaporate into the atmosphere. After printing, the solvent evaporates leaving the resin binding the pigments to the paper surface. The drying rate can be

speeded up with the aid of heat or air currents. Evaporation plays an important role in the drying of heatset web offset inks.

4. Heatset mechanism. This is similar to the quickset mechanism except that the low viscosity distillate is mostly separated from the rest of the ink by evaporation with the aid of heat from an oven. A small amount of distillate also penetrates into the pores of the paper. Drying is then completed by oxidation/polymerization. In brief, heatset drying is achieved by evaporation, penetration and oxidation/polymerization.

## **2.4 BACKGROUND ON FOUNTAIN SOLUTION.**

The two major functions of the fountain solution are to keep the non-image areas clean and free from ink and to render the minimum amount of water necessary to obtain a clean print and therefore help to maintain ink and water balance.

When image and non-image areas are produced on a plate, they will have a thin layer of gum arabic on the surface to make the plate surface extremely hydrophilic. If the plate could remain in this condition, it would be possible to apply only water onto the non-image areas. However, because gum arabic is easily worn away by the contact of roller and blanket, it is necessary to apply a desensitiser in the fountain solution to maintain this hydrophilic non-image area. Therefore, it is the desensitiser which is the main active component of any fountain solution.

Besides the desensitisers which all fountain solutions contain, other components are incorporated to improve their performance. A pH buffer is added to maintain the pH of the fountain solution. Some preservatives are added to prevent the growth of bacteria or fungi. One special additive is used to prevent the spreading of an oil film from the ink over the water film on the non-image areas. Otherwise, when the press is stopped and the moisture evaporates away, the oil film will then deposit on the non-image areas tending to print as a scum when printing is resumed.

As water is the main component in the fountain solution, its general function is to render the non-image areas inaccessible to ink. It also has some specific functions: to establish the boundaries of non-image areas, to

replenish gum to the corroded sites and to act as a medium to dissolve and transport dissolved chemical components to the plate surface.

Alcohol is known as one of the most effective wetting agent in offset lithographic process. Isopropanol is the most widely added alcohol to the fountain solution at a level of between 10 and 15 percent [Rosos, 1990]. Isopropanol dissolves in water fast and homogeneously and the functions of the alcohol are as follows:

1. To lower the surface tension of water to induce a more rapid spreading and better wetting of the non-image areas [Banks, Smith & Charlesworth, 1968; Lindqvist, Karttunen & Virtanen, 1982]. Surland [1980] stated that adding 25% isopropanol in fountain solution reduced its surface tension down to 32 dynes/cm. Others reported that an addition of 5% isopropanol to tap water lowers the surface tension of water from 75 to 50 dynes/cm [K+E technical information]. A fountain solution with alcohol will produce a thinner, more consistent film than one without alcohol.

2. Because alcohol evaporates readily from the plate surface, this helps to reduce emulsification of ink into fountain solution, obtain quicker ink and water balance, easier maintenance of this balance, and improved drying of the inks. Furthermore, there is also less water transferred to the blanket which subsequently reduces the water transferred to the paper inducing less difficulties.

3. Isopropanol increases the viscosity of fountain solution, therefore improving the transfer of fountain solution in the dampening system.

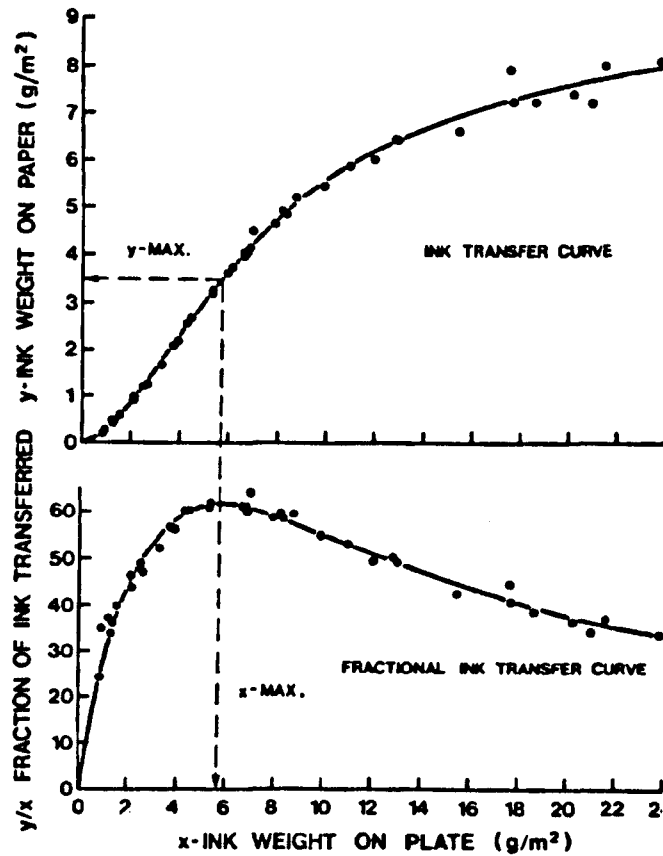
## **2.5 INK AND PAPER INTERACTIONS.**

As far as paper is concerned, the quality of a print can be related to the distribution of the ink in the paper and on the paper surface. This distribution is determined by the properties of paper and ink and their interactions in a printing process. Ink and paper interactions can be considered in 2 stages: in the printing nip (during the impression) and post-nip interactions.

### **2.5.1 In the (printing) nip interactions - Ink transfer.**

The first process in the interactions is the transfer of ink to the paper in the printing nip. The aspect of particular interest throughout this study is that of printing a single colour where the ink transfers to an uninked paper surface. The paper passes through the nip between the blanket and the impression cylinders in approximately one millisecond at full commercial printing speeds [Williams, 1988]. The ink transfer is accomplished by means of a specific amount of pressure applied at the point of contact between the blanket and the paper. Without this pressure, there would be no transfer.

In the nip, the applied printing pressure not only compresses the porous structure of the paper but also hydraulically impresses a portion of the ink into these compressed pores in the paper surface. This ink is considered to be immobilized. At the nip exit, cavitation and filamentation take place in the ink film. As the cylinders continue to move apart, the cavities expand and the ink filaments stretch. The ink transfer is completed with the fracture of the lengthening ink filaments.



**Figure 2.5 An S-shaped ink transfer characteristics curve.**

The ink transfer characteristics curve is an S-shaped curve as shown in Figure 2.5. The shape of the curve can be explained as follows [Askew, 1969; Mangin *et al.*, 1982]. With a sufficiently thin ink film, there is incomplete contact between the paper surface and the ink. This contact increases with increasing ink film thickness causing the fraction of ink transferred to the paper to increase. Ink is immobilized in the compressed pores of the paper and the remaining free ink film splits to transfer some constant fraction to the paper. The amount of immobilized ink increases as ink film thickness is increased up to a maximum value that depends on the surface porosity of paper under printing pressure. After the maximum amount of immobilised ink is obtained, the free ink film predominates. The



split fraction was found to decrease as the thickness of the free ink film increased [DeGrâce & Mangin, 1988]. Consequently, the overall fraction of ink transferred decreased.

The most well-known ink transfer equation was derived by Walker & Fetsko [1955]. This equation shows the amount of ink transferred (y) as a function of the amount of ink on the plate before the impression (x). Both amounts of ink are measured either in  $g/m^2$  or  $\mu m$ . In case of ' $\mu m$ ' the term 'ink film thickness' is used instead of the amount of ink per unit area. At low ink film thickness, the relation becomes

$$y = A [ bB + f(x-bB) ] \dots\dots\dots(3)$$

where

A is the coverage function =  $1 - e^{-kx}$

B is the immobilization function =  $1 - e^{-x/b}$

k is a constant related to the printing smoothness of the paper

b is the acceptance or immobilization capacity of the paper surface  
for the ink during impression

f is the fraction of the free ink transferred to the paper

The power function  $e^{-x}$  approaches zero as x increases. Thus at high ink film thickness, A and B approach 1. Both power functions are eliminated and the equation reduces to

$$y = b + f(x-b) \dots\dots\dots(4)$$

Equation 4 states that when there is sufficient ink to contact all parts of the paper surface, ink transfer is the sum of the two quantities: the

maximum capacity of the paper surface to immobilize the ink and a constant split fraction of the remaining ink on the plate.

Evaluating the three transfer constants 'k, b and f' provides some basic information on the printing properties on the paper and ink. For example, the constant 'k' which indicates how quickly full contact is achieved between the paper and the ink film, is used as a measure of the smoothness of the paper under printing conditions.

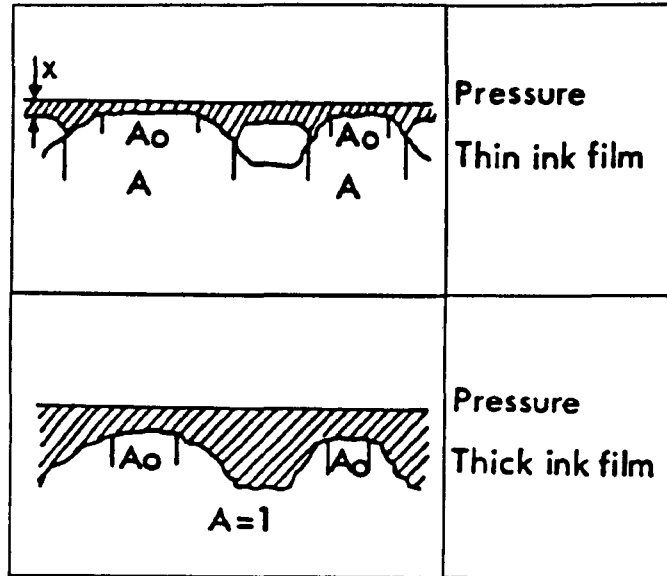
It was found [Walker & Fetsko, 1955] that the constants derived from ink transfer curves were influenced by paper and ink properties and printing conditions. All three constants increased with increasing printing pressure and decreasing printing speed. Moreover, the constant 'k' increased with increasing paper smoothness and both 'b' and 'f' increased with decreasing ink vehicle viscosity.

The ink transfer process can be summarized into three partly independent phenomena: contact between the ink film and the paper surface in the printing nip, immobilization of the ink in the surface pores of the paper and the splitting of the remaining portion of the ink film. It will be seen that the complex structure of paper creates many effects on these ink transfer components. Using a contact smoothness measurement, it was found that uncoated papers normally consist of 5-15% fibrous solid area and 15-25% for coated papers [Sear *et al.*, 1954]. The major area of the surface consists of voids or depressions. In addition, pore spaces form a large amount of the volume within the paper. Most of the pore spaces connect with each

other to form complicated three dimensional channels through the sheet, capable of drawing in liquids by the action of surface tension.

*Ink/paper contact.* The contact between the ink film and the paper surface is determined by the printing smoothness and the compressibility of the paper. Printing smoothness is referred to as the completeness of contact between the paper's surface and the ink film on the printing plate or blanket under printing pressure [Bureau, 1982]. Paper is a compressible material which means that its volume changes under pressure; the extent of this compressibility depends on the fraction of voids in the paper and the stiffness of the fibre network. In addition, the pressure on the liquid ink in the printing nip also increases the contact.

*Ink immobilization.* Karttunen, Kautto & Oittinen [1971] modified ink transfer models by introducing a new smoothness parameter ( $A_0$ ) which is the flattened fraction as illustrated in Figure 2.6. They explained that when a paper is compressed in a printing nip, there is a portion of the surface that lies parallel to the surface of the plate or the blanket. This is called the flattened fraction ( $A_0$ ) which can be covered by an ink film of any thickness. The total surface area ( $A$ ) in contact with the ink film under compression can be considered as  $A_0$  and the recess area is therefore  $A - A_0$ . The new model assumed that the liquid ink is immobilized only in the recess area and not in the flattened fraction. In other words, the ink is only split but not immobilized in this flattened fraction during the ink transfer process. Schaeffer, Fisch & Zettlemoyer [1963] interpreted the transfer parameter 'b' to include only the ink immobilized during impression where the paper is being compressed in the nip.



**Figure 2.6 An illustration of paper surface during impression.**

Karttunen [1976] proposed that most of the decrease in total pore volume in the printing nip occurs in the pores between the fibres not in the fibre lumens. The pore size of the paper in comparison with the pigment particle of the ink determines whether the ink, as a whole, can be hydraulically impressed. The pore volume under printing pressure determines the amount of immobilized ink.

Banks [1965] and Lepoutre, DeGrâce & Mangin [1979] explained that the penetration of ink into paper during impression occurred under two forces: the force of hydrodynamic pressure developed in the nip and the capillary forces. Lepoutre *et al.* derived equations; based on Hagen-Poiseuille and Laplace equations assuming a zero contact angle; to calculate the volume of ink penetration per unit area ( $V$ ) during this short impression time ( $\theta$ ) by these two forces.

$$V = (\epsilon R / 2\tau)(P\theta/\mu)^{1/2} \dots\dots\dots(5)$$

$$V = (\epsilon/\tau)(R\sigma\theta/2\mu)^{1/2} \dots\dots\dots(6)$$

where

$\epsilon$  is void volume fraction or porosity

$R$  is average pore size

$\tau$  is tortuosity (the ratio of the length of the pore to its  
end-to-end distance, e.g., the coating thickness)

$P$  is hydrodynamic pressure

$\sigma$  is surface tension of the ink

$\mu$  is ink viscosity

Olsson & Pihl [1954] and Banks [1965] showed that the printing pressure is much larger than the capillary force and they concluded that the capillary forces are negligible during impression. Many researchers have also found that the penetration rate of an ink into paper during impression in a printing nip is faster than the penetration after printing [Coupe & Smith, 1956; Hsu, 1962]. Oittinen & Lindqvist [1982] indicated that printing pressure is the primary cause of ink penetration inside the nip whereas capillary action plays an increasingly important part outside the nip. Coupe & Hsu [1960] studied penetration of varnishes and inks into paper and concluded that penetration during impression initially increases with increasing ink film thickness and is dependent on impression pressure, impression time and ink viscosity. Eventually it reaches a limiting point and becomes independent of film thickness.

Schaeffer *et al.* [1963] and Lyne & Aspler [1982] showed that under the same printing conditions, the ink immobilization of uncoated paper is

much higher than that of coated papers. The reason for this is that the mean pore size in the surface of coated papers is smaller than in uncoated papers [Lepoutre *et al.*, 1979]. The results indicate why heatset inks are often used in coated papers in order to prevent set-off.

In considering the ink's influences, ink rheological properties have little effect on the ink immobilization 'b' with coated paper. However, with uncoated papers, increasing viscosity decreases the ink immobilization in the nip [Schaeffer *et al.*, 1961].

At the nip exit, the ink which is impressed into the paper under hydrodynamic pressure cannot be sucked out again by the lower pressure splitting force [Hultgren, 1971]. Moreover, it was observed that the penetration rate of ink into the paper is higher than the rate of recovery of the penetrated layers of the compressed paper. As a result, as soon as the paper regains its original size, the created space is filled with the ink drawn from outside [Hsu, 1961].

*Ink film splitting.* Ink splitting occurs in the total coverage area (A). Wetting, spreading and adhesion are important to ink film splitting. The first requirement for splitting is wetting and spreading of the ink over the surface of the paper. In order to achieve spreading of the ink on the paper, the contact angle between the ink and the paper must be zero which occurs only when the ink completely wets the paper. The criterion for this is that the surface energy of the paper ( $\gamma_p$ ) must exceed the sum of those of both the ink ( $\gamma_i$ ) and the interfacial tension between the paper and the ink ( $\gamma_{ip}$ ). Under other conditions, spreading does not occur.

$$S = \gamma_p - \gamma_i - \gamma_{ip} \dots\dots\dots(7)$$

The second requirement, in order that splitting can occur in the ink film, is that the adhesion strength of the ink with the paper must be stronger than the cohesive strength of the ink itself. The work of adhesion (W) between the ink and the paper is defined as

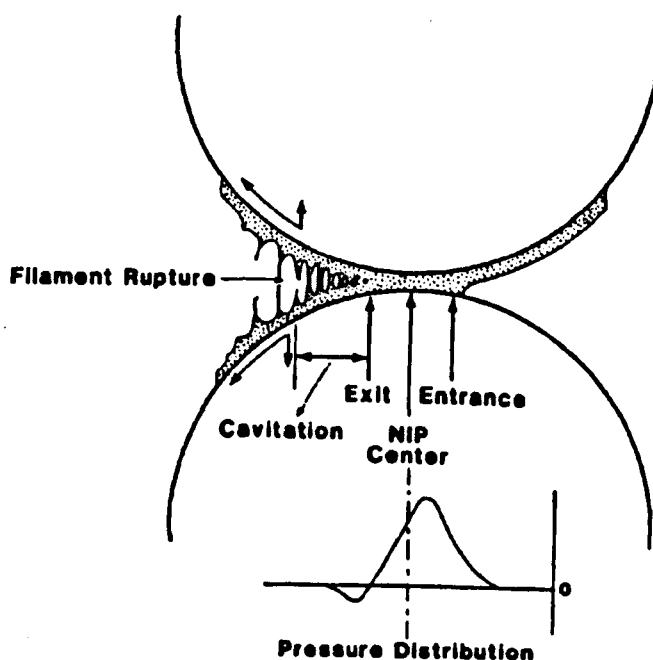
$$W = \gamma_p + \gamma_i - \gamma_{ip} \dots\dots\dots(8)$$

The adhesion of the ink film to the paper is enhanced by factors such as the roughness of the paper and the polarity of both the ink and the paper. Increasing the roughness of the paper provides greater potential areas of contact for adhesion to take place. If the ink and the paper contain polar groups, the negative charges in the ink film may orient themselves close to the positive charges on the paper surface and *vice versa* to improve the adhesion.

Banks & Mill [1954] studied the ink film splitting mechanism at the exit of the nip formed by two rollers. They considered this mechanism to involve cavitation, filament formation, filament elongation and filament rupture. Using a high speed photographic technique to observe ink filaments, DeGrâce, Dalphond & Mangin [1992] suggested that ink film splitting in the printing nip, where the ink is brought into contact with an uninked paper surface, also follows the same sequence. They explained the basic mechanism of ink film splitting as follows.

As the ink emerges at the nip exit, there is a sudden reduction of pressure in the ink. This reduction of the pressure causes dissolved air in

some ink components to emerge and form cavities within the ink film [Myers, Miller & Zettlemoyer 1959]. As the two cylinders continue to move apart, the cavities expand vertically and ink filaments are formed between the cavities. The ink filaments elongate and become thinner. Eventually, ink film splits as the ink filaments rupture. Figure 2.7 shows ink film splitting at a nip exit.



**Figure 2.7 Ink film splitting at a nip exit.**

Theoretically, ink film will split 1:1 (50%) between two rollers moving at the same speed. However, if the rollers move at different speeds, ink film splitting will always yield more ink to the faster roller [Fetsko, 1989]. Many researchers reported that the free ink splits asymmetrically [e.g., Walker & Fetsko, 1955; Zettlemoyer & Fetsko, 1956]. In addition, some have made an attempt to explain the origin of this asymmetric splitting. Fetsko [1958] suggested that air bubbles from a porous paper could enter the ink film and



cause ink splitting near the paper surface. Taylor & Zettlemoyer [1958] proposed that there was a region of low viscosity ink film closer to the paper surface. The cavity growth and the ultimate film splitting proceeded in the direction of low viscosity, that is towards the paper. DeGrâce & Mangin [1984, 1988] explained that surface asperities on the substrate surface and air entrainment at the nip entrance may act as nucleation sites for cavitation and cause subsequent ink film splitting to occur near the substrate surface, probably at the very tip of the surface asperities.

Karttunen [1976] reported that paper properties have little effect on splitting. The most important factors which influence the splitting constant ' $f$ ' are printing speed and ink viscosity. The splitting constant ' $f$ ' varies from about 0 to 0.5. High printing speed and high viscosity ink give low values of ' $f$ ' whereas slow printing speed and low viscosity ink give high values of ' $f$ '.

The force required to split an ink film is termed 'tack'. Banks & Mill [1954] stated that tack is a consequence of cavitation and corresponds to the maximum negative pressure occurring in the nip. Two important factors involving tack are cavitation and ink rheology. Hoffman & Myer [1962] analysed a cavity expansion within an oil and found that the contribution of surface tension to the force required to separate a liquid film (tack) is about 7% of that of the viscosity. Banks [1965] explained that tack forces will be smaller when cavitation occurs in the ink film. The effect of ink viscosity on the splitting constant ' $f$ ' was also observed by Schaeffer *et al.* [1961]. He found that the portion of the free ink film which splits towards the paper also decreases with increasing ink viscosity.

Pigmentation increases the number of cavities formed (Myer *et al.*, 1959). Lyne & Aspler [1982] stated that suspensions cause cavitation more readily than fluid vehicles. Large pigment particles will initiate cavitation sooner, thus the tack force is smaller. Conversely, smaller pigment particles will cause cavitation to occur later. This results in finer filaments which in turn improve print quality by improving the uniformity of ink transfer.

Ink transfer to paper under the conditions that occur in a printing nip was studied by DeGrâce & Mangin [1984]. They examined the effects on ink transfer of substrate properties such as surface free energy, roughness and porosity and of press conditions such as printing pressure and speed. They found that the surface free energy did not affect ink transfer. A greater amount of ink was hydraulically impressed into a porous substrate such as newsprint than into a non-porous substrate at the same level of roughness. Moreover, printing pressure was found to affect only the immobilized fraction while printing speed has a marked effect on both the immobilized and the split fractions. This was also found by Schaeffer *et al.* [1963] who explained that for coated papers, the rheological properties of the ink determine the 'b' value whereas printing conditions as well as paper influence 'f' more than 'b'. In the case of uncoated papers, both 'b' and 'f' are significantly influenced by printing conditions.

Lepoutre & DeGrâce [1978] and Lepoutre *et al.* [1979] investigated the effects of coating structures with different absorption properties on ink transfer, print density and print gloss. Ink transfer characteristics were found to be independent of the absorption properties of the coatings but were only a function of their surface roughness. Print density and gloss increased

with the amount of ink deposited on the coatings. They concluded that no ink absorption into the coating occurs during the impression but the immobilized ink is contained in the surface crevices. Furthermore, uneven print density and gloss are mainly influenced by the variations in thickness of the ink film deposited, these being related to the uneven surface profile and compressibility of the coated papers.

Apart from surface roughness and porosity properties, oil absorbency and water absorbency are two different properties which are of importance in offset lithographic printing. Ink films transfer from one surface to another by splitting. This splitting creates a split pattern which remains for a certain time until the ink levels out. The oil absorbency of paper is important at this stage because if the paper absorbs thin oil too fast before the leveling out is completed, it will result in roughness of the ink film. Water absorbency is very important on multicolour printing where the dampened non-image area of the first unit may become the image area of the second unit. If the second ink contacts the fountain solution instead of the paper surface, ink repellance may occur.

### **2.5.2 Post-nip phenomena - Ink drying or consolidation of the ink film.**

Post-nip interactions that occur between the transferred ink film and the paper are normally referred to as 'ink drying'. Ink drying goes through two phases: ink setting and ink hardening.

Ink setting mostly involves penetration into the paper or evaporation (and leveling) of the ink vehicle causing the ink to 'gel' or 'set'. The ink film is

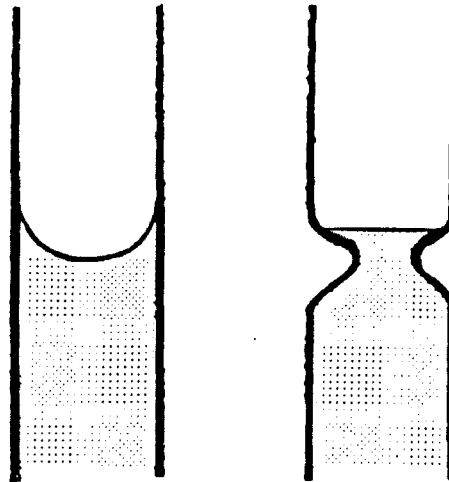
not completely dry at this stage but is sufficiently dry to be handled mechanically. Adhesion of the ink film to the paper is influenced by the degree of penetration. Ink hardening involves oxidation/polymerization of the resin into a hard film that adheres to the paper surface.

*Penetration.* The penetration of ink into paper which depends on the relative size of the ink pigments and of the surface pores has been studied by several researchers [e.g., Coupe & Hsu, 1960; Hsu, 1962; Levlin & Nordman, 1967; Larsson & Trollsas, 1967, 1969; Christensen, 1967]. The mechanism can be explained as follows. In the nip, ink penetrates as a whole into the larger pores and depressions on the surface of the compressed paper. After the nip, the paper recovers from its compressed state, most of the pigments which have penetrated to a depth of 20-30% of the paper thickness remain in their penetrated layers [Levlin & Nordman, 1967]. The vehicle moves further into the paper under capillary action, leaving the large pores on the paper surface open. The surface pores in paper are modelled as being conical.

DeGrâce & Dalphond [1989] observed that pigment penetration ended within 3 seconds after printing. This implied that pigment particles penetrated into the compressed surface through hydraulic impression and through aspiration of the paper after the nip.

The capillary force depends on the pore system constituted by the fibres and pigments. A condition for the capillary force to act on the vehicle is that the vehicle completely fills up the pore, i.e., the vehicle does not move along the fibre surfaces only. The irregularities in the capillaries will decrease the radius of the meniscus and thus retard penetration. Figure 2.8

(right) shows the penetration of liquid into an irregular capillary. When the liquid has penetrated to a point where the diameter increases quickly, the meniscus becomes flattened and the capillary action ceases.



**Figure 2.8 Penetration of a liquid into an irregular capillary.**

Penetration of a liquid under capillary pressure in a horizontal capillary where gravity can be neglected is described theoretically by the Lucas-Washburn equation [Washburn, 1921]:

$$l = (r\gamma\cos\theta / 2\eta)^{1/2} t^{1/2} \dots\dots\dots(9)$$

where

$l$  is penetration depth after time  $t$

$r$  is effective pore radius

$\gamma$  is surface tension

$\theta$  is contact angle

$\eta$  is liquid viscosity

This Lucas-Washburn equation is a simple equation to model the rate of capillary penetration of non-swelling liquids such as oil-based inks. However, it is not suitable for the penetration of liquids such as water or other polar liquids into paper. One of the reasons is that swelling of the fibres as a result of water penetration affects the penetration rate.

Olsson & Pihl [1954] developed an equation of the Lucas-Washburn type to calculate the penetration depth (T) of vehicle into paper.

$$T^2 = (V/\pi r^2)^2 = [(2r\gamma\cos\theta + Pr^2)/4\eta] t \quad \dots\dots\dots(10)$$

where

V is volume of vehicle that has penetrated at time t

r is mean effective pore radius

$\gamma$  is surface tension of vehicle

$\theta$  is contact angle between vehicle and structure

P is pressure on vehicle

$\eta$  is viscosity of vehicle

t is penetration time

Equation 10 can be used for both nip penetration and post-nip penetration. For example, when  $P = 0$ , the post-nip penetration depth of vehicle by capillary force only can be calculated.

The dependence of liquid penetration on coating absorption properties was examined by Lepoutre [1978]. He found that the absorption rate decreased exponentially with decreasing porosity. The reduction in the porosity of a coating as a result of the calendering process also decreases the oil penetration rate.

The progressive penetration of the vehicle results in an increase in the pigment concentration in the ink film. Capillaries are created between pigment particles within the concentrated ink film and begin to exert an external resistance to the flow of ink which is subsequently equal to that in the paper [Bristow & Bergenblad, 1992]. That is the ink penetration process continues until an equilibrium between capillary forces in the ink film and in the paper is obtained. Thereafter the vehicle is retained by the pigments. The retention of ink pigments and high molecular weight resin is a function of the absorbency of paper and is known to play a part in ink drying for coated papers.

The penetration of pigment particles in the ink into papers is often obstructed by the filtration mechanism of the paper structure. Coupe & Smith [1956] studied this phenomenon and stated that in all cases, the vehicle has penetrated deeper into the paper than the pigments and that the extent of this filtration depends on the pore size. Papers having small pores such as coated papers will show greater filtration than one having large pores such as uncoated papers. Hattula & Oittinen [1982] used K&N ink to examine the ink penetration into a coating structure with very small pore sizes. They reported that very little pigment penetrated whereas the solvent penetrated to a greater extent. As a result the coating structure showed a gradient in pigment and vehicle concentration. This has also been observed by Larsson & Trollsås [1967]. They introduced a method of splitting a printed sheet into layers parallel to the plane of the sheet and found that the pigments were distributed with decreasing concentration in the transverse direction from the printed side of the paper. However, Banks [1975] stated that there is no filtration of pigments for uncoated papers

whereas the coating pores of coated papers are smaller than the pigment particles which are therefore filtered out.

Larsson & Sunnerberg [1972] studied these two phenomena: penetration and filtration of the ink. They found that penetration occurs mainly within the first minute after printing. Filtration occurred over a much longer period of time and changes in pigment concentration occurred quickly, during the first few minutes. They believed that equilibrium was reached after twenty-four hours. Sørensen [1982] measured changes in tack of the ink film with time. The results supported the suggestion that pigment concentration increases as the ink vehicle penetrates into the coatings.

The uniformity of vehicle penetration into the paper after the nip is influenced by the sheet formation which in turn affects the surface characteristics of the ink film remaining on the surface. Continued vehicle penetration may cause the remaining pigment layer to follow the surface contour of the paper resulting in an uneven ink film on the surface [DeGrâce & Dalphond, 1989]. The uniformity of pigment distribution is thus determined by the roughness of the uncompressed paper.

### **2.5.3 Ink and paper interactions in offset lithographic process.**

As has been discussed earlier, the offset lithographic process involves ink and fountain solution interactions prior to ink and paper interactions. This aspect has to be considered in addition to the general understanding of ink and paper interactions. Surland [1980, 1983] explained that the dynamic printing of offset lithographic process involves emulsion formation and



emulsion breakdown, which can be written in a parametric equation as follows.



where W is fountain solution or water and O is ink.

He developed a method to determine the rate of the emulsification of fountain solution into ink. The emulsification test provides a P-curve of the progressive change between these opposite vectors.

$$P = P^+ / P^- \hspace{1cm} \text{.....(12)}$$

where P<sup>+</sup> is the emulsion formation rate and P<sup>-</sup> is the emulsion breakdown rate. The rate of emulsification of fountain solution or water into ink can be used as an indication of press performance. Chou, Fadner & Bain [1987] stated that the success of offset lithographic printing in relation to ink and water interactions depends on the emulsification capacity and the emulsion rate of fountain solution into the ink.

If water only is used as the fountain solution, any polar components in the inks can form a film onto the non-image area which has already been covered with water. To prohibit such film formation, Banks *et al.* [1968] found that non-ionic surfactants or alcohols are necessary additives in the fountain solution. Bock [1969] examined several functions of alcohol used in the fountain solution and he found that when isopropanol below 12% by volume is used, the water feed rate rises rapidly. These high water feed rates can cause a number of printing problems such as softening of the paper coating, and expansion of paper or snowflaking. Surland [1980] showed that

the addition of isopropanol into fountain solution improves the emulsification characteristics that in turn improves press performance. The study of Douw & Blokker [1989] indicated that an addition of only 1% isopropanol to an ink caused the viscosity of ink to reduce by 50% which in turn improves the emulsification. Braun [1985] reported that the emulsified droplets in the emulsion ink are much finer in the presence of isopropanol.

Two terms are usually used to express the emulsification results: water content and water pick up. Water content is the relative amount of water or fountain solution in an emulsified sample. It is expressed as  $\text{g H}_2\text{O} / 100 \text{ g emulsion}$ . Water pick up is the amount of water relative to that of the ink in an emulsified sample. It is expressed as  $\text{g H}_2\text{O} / 100 \text{ g ink}$ . Cunningham & Moore [1984] postulated that an ideal offset ink should absorb water or fountain solution approximately 21% by weight under high shear condition without adversely changing its physical properties. Chou & Cher [1989] pointed out that with increasing water content, the stability of an emulsion will decrease.

Many workers have studied the rheology of emulsion inks. Emulsification of fountain solution in offset inks was found to reduce the viscosity but increased the yield value and the shortness, which is the ratio of yield value to viscosity, of the ink [e.g., Lavelle, Schaeffer & Zettlemoyer, 1969; Bassemir & Shubert, 1985]. However, this trend was not always the case since the emulsification behaviour varied from system to system, i.e., with different materials. Emulsification enhances ink transfer to porous substrates but had no effect on non-porous substrates.

Using infrared analysis to determine the water content of offset inks at various stages of printing, Cartwright & Harden [1965] found that the water content dropped off from 1% on the blanket to 0.5% on the print due to the action of evaporation. MacPhee [1985] also pointed out that most of the aqueous component of fountain solution is evaporated in the inking system. Therefore the composition of fountain solution certainly changes with time. By the time it reaches the plate or the blanket, the concentration of the volatile components have decreased. In addition, Karttunen, Lindqvist & Virtanen [1988] employed neutron activation analysis to determine the amount of ink and fountain solution transferred to paper. They found that the amount of fountain solution transferred to paper is higher in the image area than in the non-image area regardless of the different types of fountain solution used. This was also found in the study of Nieminen [1992]. The amounts of fountain solution transferred from the non-image areas at usual feed levels were less than  $0.5 \text{ g/m}^2$  whereas the amounts transferred to image areas in all cases were greater than  $0.5 \text{ g/m}^2$ .

The influences of these phenomena on the characteristics of a print have been studied mostly in relation to print density and print gloss. It is known that gloss depends on smoothness or roughness. Interactions between water and paper in offset lithographic printing may result in an increase in macroroughness as found in paper coating process [Skowronski & Lepoutre, 1985]. The effects of water on paper smoothness were also studied by Hansuebsai [1989] who found that water decreased paper smoothness in all cases. Koniecki *et al.* [1983] found that print density and print gloss obtained with high water uptake inks are slightly higher than those obtained with low water uptake inks. Ginman & Visti [1971] reported that for web offset

printing, the main factor which causes a decrease in gloss is moisture. Heating plays a minor role. The change in gloss is accompanied by a change in smoothness.

It can be seen that there has been much study of ink transfer phenomena, in particular, ink penetration and ink film splitting; and of the emulsification behaviour and the resultant properties of the emulsion inks. Little work, however, has been concerned with the effects of these phenomena on print quality. The qualities of interest mostly evaluated were print density and print gloss. It is attempted, in the present work, to study the ink/paper/print quality relationships by applying methods to evaluate print smoothness. This aspect has not been widely examined. This is probably because the achievement of print smoothness involves many factors in the printing process, as previously discussed. However, it is this quality of '*smoothness*' to which many other print qualities, such as print gloss and print mottle are related. The present study thus arose from the need to develop such understanding.

## **2.6 SURFACE CHARACTERISTICS CONSIDERATION AND MEASUREMENTS.**

The preceding section has dealt with ink and paper interactions and also gave particular attention to the offset lithographic process; where fountain solution is present and interacts with the ink and paper. All of the phenomena in the nip and after the nip are of importance to the quality of a print.

Print quality (or the quality of a print) is the degree to which the appearance characteristics of the print approach those of the desired result regardless of how it was produced [Walker & Carmack, 1964]. High print quality is dependent on the desired results. Print quality may not be a single property but a composite of many contributing factors whose relative importance varies between jobs. Therefore, a print as a result of any printing process needs a clear statement of which quality factors are desirable. Print quality factors are mostly classified in relation to printability.

Printability of a surface is the degree to which its properties enhance the production of high quality prints in a particular process. It is apparent that printability of a surface includes all properties that influence the quality of the prints made on it. From the section on ink and paper interactions it can be seen that one of the most important factors affecting print quality is the surface structure of paper [DeGrâce & Mangin, 1984; Walker & Fetsko, 1955]. This is usually referred to in the literature as surface smoothness (or roughness).

Several different methods have been used for the determination of this surface structure. They can be divided into direct and indirect methods. Most indirect methods measure physical properties which are controlled by surface structure over a relatively large sample area and do not give detailed information. Direct methods measure geometric profiles therefore giving more detailed information.

### 2.6.1 Direct method.

*Surface profile measurement.* A qualitative method is simply to observe the magnified surface using instruments such as a magnifying glass or an electron microscope. Attempts have been made to describe the surface structure quantitatively. A more useful method is surface profile measurement. This method mechanically scans the surface with a contacting stylus and records the vertical movement of the stylus arm. One example of this instrument [Higgins *et al.*, 1973] scans the surface of a paper sample over a 50 mm length which is divided into 1 mm intervals with 120 readings per interval. The readings are used to calculate the r.m.s. (root mean square) ( $\sigma$ ) of the vertical displacement as the roughness of the surface.

$$\sigma = \left[ \frac{1}{l} \int_0^l Z^2 dx \right]^{1/2} \dots\dots\dots(13)$$

where

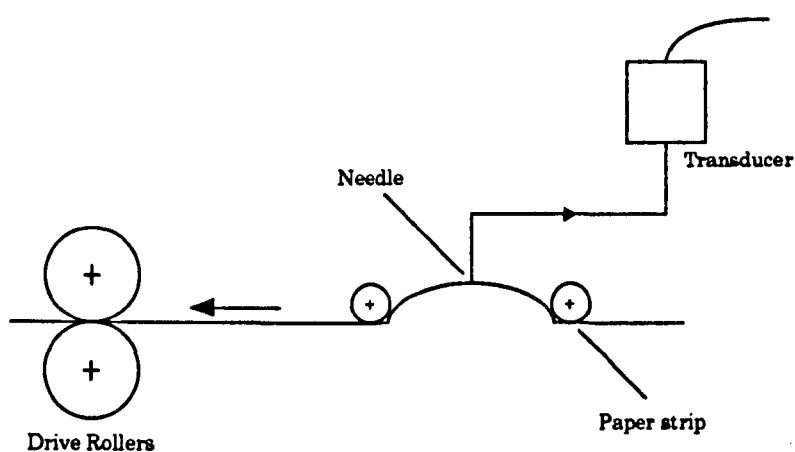
$l$  is a profile length

$Z$  is a height measured from the mean line

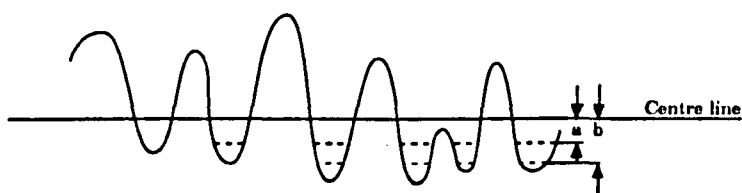
The Talysurf 10 is a commercial instrument modified to involve both the vertical and horizontal directions and produces a centre line average (c.l.a.) of the profile as a measure of surface roughness. The centre line

average value is determined as the average of the departure of the profile from its centre line throughout the sampling length. The centre line is defined in BS 1134, Part I : 1972 as a line so placed that the sums of the area contained between it and those parts of the profile which lie on each side of it are equal.

Another study was done by Ginman, Makkonen & Nordman [1973] to characterize surfaces of newsprint and magazine paper. They used a specially built profile tester as shown in Figure 2.9a. From the profile curve (Figure 2.9b), they calculated the profile roughness which is defined as the cumulative width of depressions at a level which is a given distance below the centre line.



**Figure 2.9a A diagram of a profile tester.**

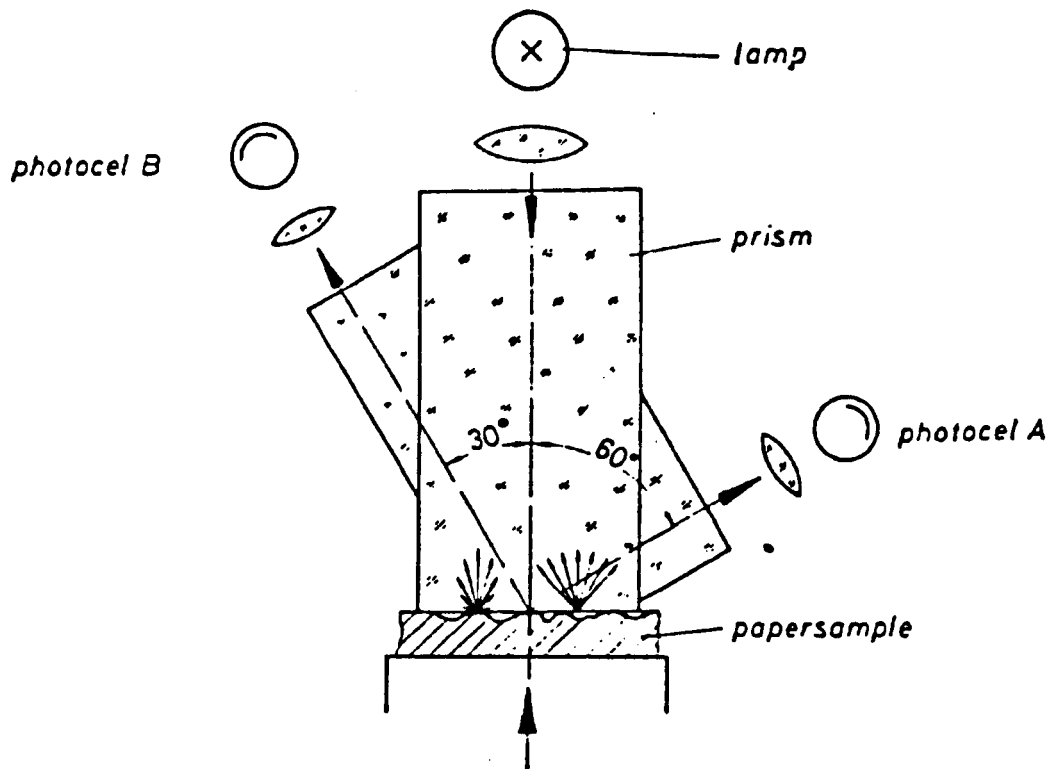


**Figure 2.9b An illustration of a surface profile.**

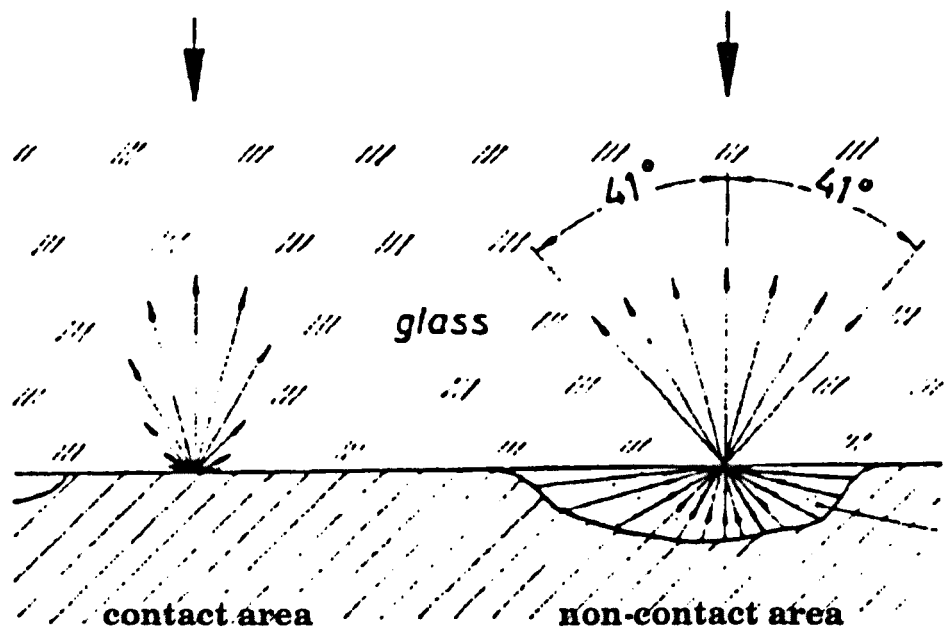
*Optical contact method.* Paper is compressible and when printed, is subjected to a printing pressure. This has an effect on the smoothness of paper. Therefore it is desirable to measure the 'printing smoothness' of paper. Printing smoothness is defined as a measure of how well full contact between the paper surface and the ink image on the plate or blanket surface is attained under printing conditions. Complete coverage of the paper surface by ink occurs close to the maximum in fractional ink transfer [DeGrâce & Mangin, 1984]. A high printing smoothness paper gives complete coverage with a minimum of ink film thickness and pressure [Walker & Carmack, 1964].

An instrument which is designed to determine printing smoothness optically is the Chapman smoothness tester [Chapman, 1954]. As illustrated in Figure 2.10, the principle of this instrument is that the sample is pressed against a flat glass prism and illuminated normally by a parallel beam of light through the prism. This incident light is partly absorbed and partly reflected back towards the glass prism. The latter is dependent on whether or not the paper is in optical contact with the glass prism. The light from the contact areas is diffusely reflected back into the prism in all directions. On the other hand, that from the non-contact areas enters the glass prism from an air medium and is refracted towards the normal according to Snell's law. The light from the non-contact areas enters the glass prism at less than the critical angle to the normal (about 41 degree from the normal). It is shown that the detected light outside the critical angle thus originates only from the contact areas whereas that detected within the critical angle will contain light from both contact and non-contact areas. These two measured values are used to calculate the Chapman printing smoothness according to





**Figure 2.10a Layout of Chapman Smoothness Tester.**



**Figure 2.10b Layout of the principle of the Chapman smoothness tester.**

$$F = \frac{MA}{B+NA} \dots\dots\dots(14)$$

where

F is the fractional contact area or printing smoothness

A is the illumination on photocell A arranged to detect light from outside the critical 41 degree cone.

B is the illumination on photocell B arranged to detect light from within the critical 41 degree cone.

M and N are the instrumental constants.

Another approach to characterize the roughness of paper under actual printing conditions was proposed by Mangin & Geoffrey [1990]. The printing roughness is calculated from the modified ink coverage function in the ink transfer equation as follows:

$$A(X) = 1 - (1 - F_0) e^{-(kx)^\gamma} \dots\dots\dots(15)$$

where

k is the smoothness parameter

F<sub>0</sub> is the flattened fraction

γ is the compression parameter

The printing roughness (R<sub>g</sub>) is given by

$$R_g = \left[ \frac{6 (1 - F_0)}{k^{3\gamma}} \right]^{1/3} \dots\dots\dots(16)$$

where k, F<sub>0</sub> and γ are obtained from ink transfer data. The proposed printing smoothness correlates well with conventional air leak methods.

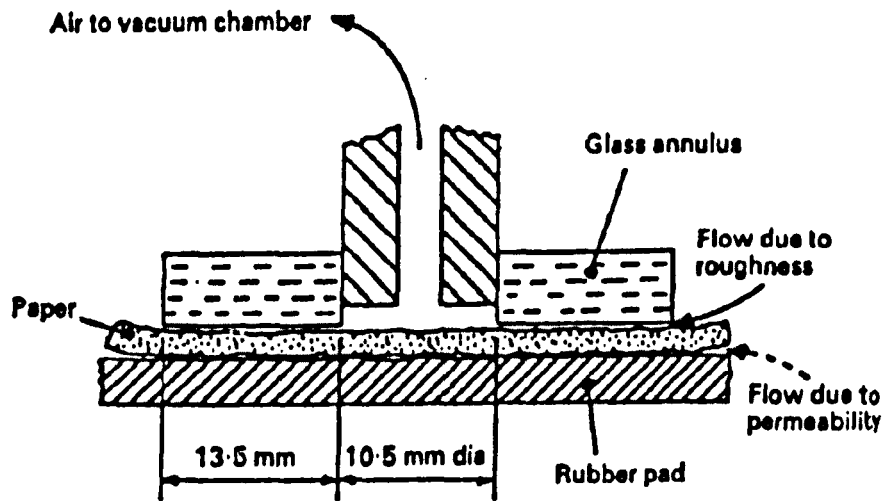
### 2.6.2 Indirect method.

*Air-leak methods.* Roughness from these methods (Figure 2.11) is determined by the rate of air-flow through voids formed between the paper surface and a flat surface.

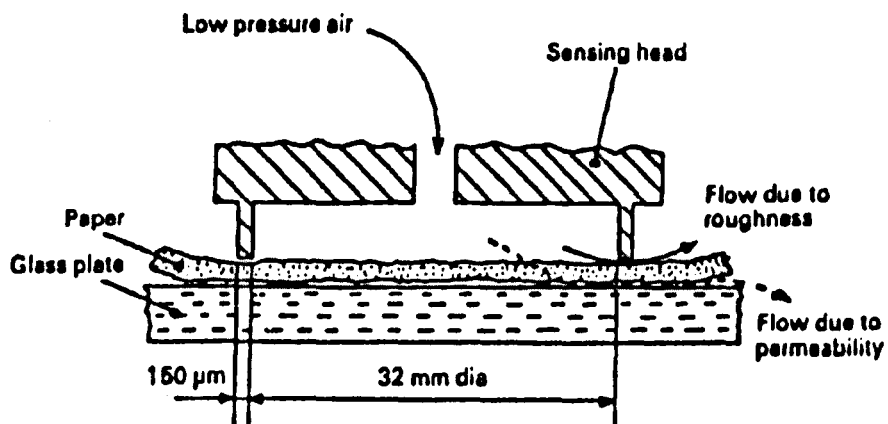
The Bekk's air leak tester was the first of the well-known methods in this group. The sample is pressed against an annular glass surface of 10 cm area and a vacuum is applied to the hole in the centre. Air flows between the paper surface and the glass. The flow rate of the air is a measure of roughness of the paper.

The Bendsten roughness tester, as described in British Standard 4420:1969, adopts the Bekk's air leak principle. The sample is pressed by a flat annular metal ring. The air at a constant pressure is supplied to the centre of this ring and then escapes between the annulus and the sample. The rate of airflow between the annulus and the sample is measured. Both the Bekk and the Bendsten tester measure roughness in units of ml/min.

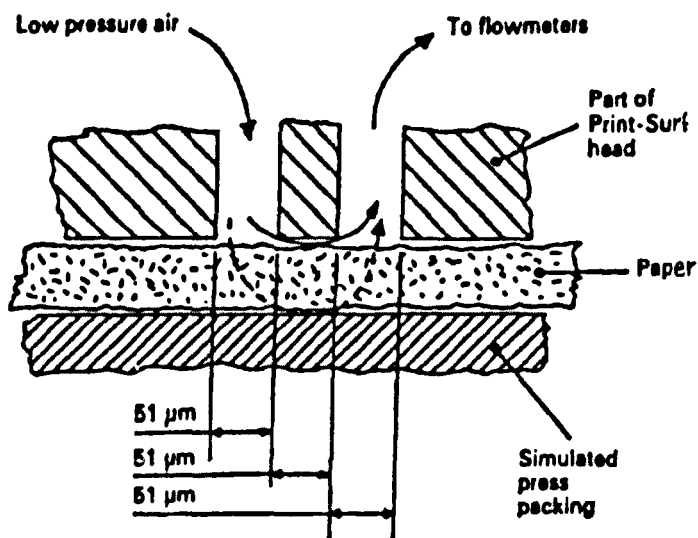
The Parker Print-surf air leak apparatus (PPS) was developed by Parker [1971]. As described in British Standard BS 6563:1985, the PPS eliminates a major error which normally occurs in other air leak methods caused by the flow of air through the whole paper. Another error, the distortion of the paper surface caused by the use of a high clamping pressure, is avoided by a better design of its sensing head. With PPS, the sample is pressed at a fixed pressure by a resilient backing against a reference plane which is interrupted by two narrow parallel slots separated by a metering



**The Bekk's air leak tester.**



**The Bendsten roughness tester.**



**The Parker Print-surf air leak apparatus.**

**Figure 2.11 Air leak instruments.**

land. Air is supplied at constant pressure to one of the slots and the rate of airflow between the surface and the metering land into the other slot is measured and thereafter is used to calculate PPS roughness. PPS instrument determines roughness in unit of  $\mu\text{m}$ .

*Fluid transfer and coverage method.* This is a method to obtain paper roughness by filling the voids on the paper surface with one material and determining the amount of that material used. Roughness in this case is the surface void volume. Hart, Verhoeff & Galley [1962] used a trailing blade device to coat paper with lanolin. This produced a reference surface level at the height of the maximum profile. The amount of lanolin used was taken as a measure of the roughness. It could be determined by several means, such as: gravimetrically, by weighing the paper before and after coating; or colorimetrically, by adding an oil-soluble dye into the lanolin, which was subsequently extracted from the measured area and the dye which remains on the surface was then determined photometrically.

Hsu [1964] suggested a criterion to distinguish between surface crevices (which constitute roughness) and pores (which do not) by considering the time taken for the liquid to flow into the paper surfaces. He postulated that the filling of surface crevices will take nearly zero time whereas filling of the pores will take some time.

Using fluids other than inks may not give a roughness identical to the printing roughness required, therefore Hsu [1964] proposed a method using ink as the medium for the determination of paper roughness. The ink was transferred to the paper on an IGT printability tester to produce a range of

film thicknesses. Various roughness parameters were determined from the diffuse reflectance data of the surfaces measured immediately after inking. They are found to correlate to some extent with other roughness methods such as the Bendsten, Chapman and Talysurf. The method concerns the whole distribution of depressions rather than an average roughness value of the sample and Hsu suggested that the depression at which the maximum frequency occurs should be taken as a surface roughness index.

All of the methods previously described have been useful to a degree but have their own particular limitations as follows.

1. Surface profile measurement gives the profile of an uncompressed surface. If the size of the stylus is larger than the roughness of the paper, it may not bottom in the crevices on this smoother surface. This results in inaccurate measurements. Making the point of the stylus finer may present further drawbacks to the method as the stylus may damage the surface. The method detects a very small area and is time consuming. For example, stylus profilometry takes from 20 minutes to 1 hour to generate a 3D map of 1 mm of paper surface whereas PPS takes only a few seconds [Mangin, 1990]. Also it is difficult to relate the profile result to actual printing performance.

2. The condition of the Chapman optical contact method is that the sample is made to contact the glass prism optically and is then illuminated. The amount of reflected light is related to smoothness. This optical contact condition in turn becomes a drawback of this method because it does not fully relate to printing. The paper only approaches the plate or the blanket within the thickness of the ink film. Since the ink at this state is quite fluid, it can transfer to the paper under the printing pressure applied.

3. Most air leak methods examine the paper surface under pressures far less than those which occur in actual printing. The roughness results are subjected to errors caused by the compressibility of the paper and by the flow of air through the paper [Ginman *et al.*, 1964; Vloodt, 1964; and Walker & Carmack, 1964; DeGrâce & Mangin, 1984;]. Thus a paper with many small voids may produce the same reading as one with a few deep voids. In addition, the result gives the roughness of paper surface in dimensions of total volume.

4. The fluid transfer and coverage methods in most cases do not give a definite figure but provide subjective comparisons between various papers. Hsu's method examines roughness under specific conditions so that the roughness obtained is that under the impression pressure of the printing process. However, the method is very time consuming.

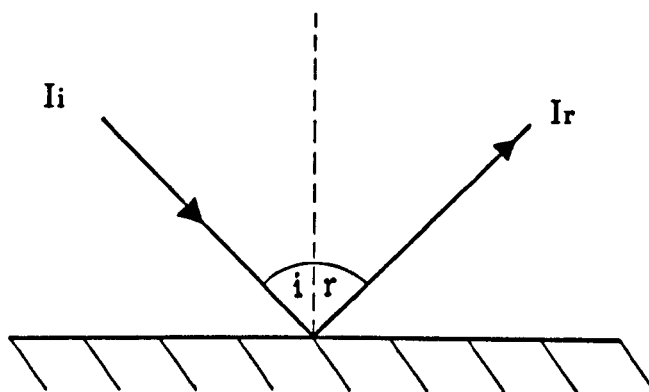
*Non-contact optical method.* This is generally a method to characterize surface structure by reflectance measurements where the light incident on the surface is partly reflected or scattered and partly absorbed or transmitted through the surface. Therefore, a knowledge of the laws governing reflectance, refraction and scattering of light is needed for a better understanding of this method.

### 2.6.3 Reflectance theory.

#### *Type of reflectance.*

1. Specular reflectance. The law of mirror reflectance states that light incident at an angle, as measured from the normal to the surface, is reflected in the same perpendicular plane on the opposite side of the normal so that the reflected angle ( $r$ ) is equal to the incident angle ( $i$ ). For a perfect mirror surface (Figure 2.12), the intensity of the reflected light ( $I_r$ ) is equal to that of the incident light ( $I_i$ ) at every specular angle.

$$r = i \text{ and } I_r = I_i \quad \dots\dots\dots(17) \text{ Law of mirror reflection}$$



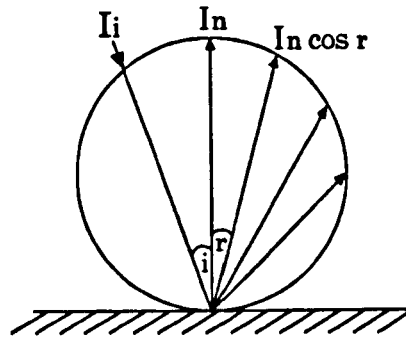
**Figure 2.12 Specular reflectance from a perfect mirror surface.**

2. Diffuse reflectance. The law of diffuse reflectance states that the intensity of the reflected light at any angle is proportional to the intensity of the incident light, the cosine of the incident angle and the cosine of the reflected angle.

$$I_r = I_i \cos i \cos r \quad \dots\dots\dots(18) \text{ Lambert's law}$$

For an ideal diffuse surface (Figure 2.13), if  $I_n$  is the intensity of reflected light normal to the surface, the intensity at any angle is given as  $I_r = I_n \cos r$  since  $\cos 0^\circ = 1$ .



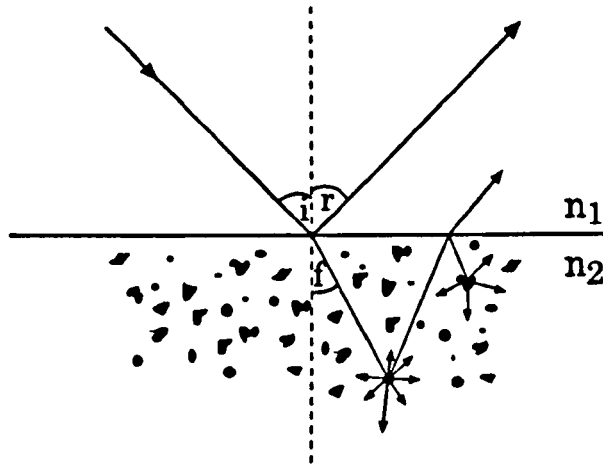


**Figure 2.13 Diffuse reflectance from an ideal diffuser.**

3. Refraction or internal diffusion. For all surfaces except a perfect mirror surface, the incident light is partly reflected directly from the surface and in part penetrates the surface to be refracted or transmitted. Figure 2.14 illustrates the reflectance and refraction from a smooth pigmented film. The incident light penetrates the surface at an angle ( $f$ ). The extent of refraction is dependent on the refractive indices of the film ( $n_2$ ) and the air ( $n_1$ ). This is Snell's law:

$$\frac{\sin i}{\sin f} = \frac{n_2}{n_1} \dots\dots\dots(19)$$

This refracted light is then either diffusely reflected from the pigmented film and re-emerges to join with externally reflected light or is reflected further for repeated refraction by pigment particles which will absorb some of the refracted light. The overall reflectance from the surface is therefore affected by multiple reflectance of the refracted light.



**Figure 2.14 An illustration of the reflectance and refraction from a smooth pigmented surface.**

*Laws of reflectance.*

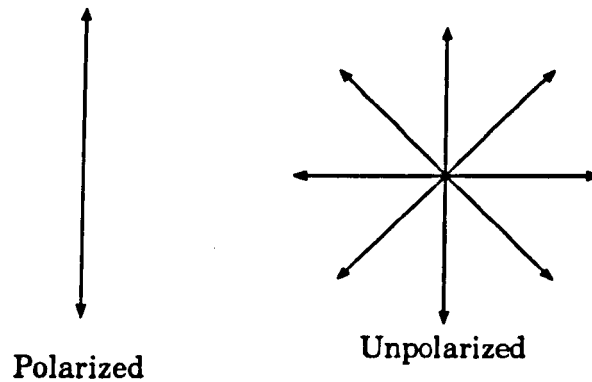
1. Fresnel law for specular reflectance. The Fresnel law applies to a surface which consists of macro sized elements compared with the wavelength of incident light. For light which is incident at an angle of zero degrees to a non-metallic surface with a refractive index of  $n_2$  in contact with a less dense medium  $n_1$ , such as air, the reflectance intensity (R) is given as:

$$R = \left[ \frac{n_2 - n_1}{n_2 + n_1} \right]^2 \dots\dots\dots(20)$$

When the incident angle is not at zero degrees, the Fresnel law states that the intensity of specularly reflected light increases with the refractive index ratio, with angle of incidence and the degree of polarization of the light (See equation 21,22).

A light wave is considered to consist of time dependent magnetic and electrical fields perpendicular to each other and vibrating perpendicularly to the direction of propagation. Unpolarized light has no preferential plane of

vibration and the plane of vibration alternates many times per second. However, there is light where such vibration remains in only one plane. Such light is called (linearly) polarized light. Figure 2.15 illustrates linearly polarized and unpolarized light. The plane of polarization usually referred to is that of the electrical field and is either perpendicular or parallel to the plane containing the incidence and reflection.



**Figure 2.15 Polarized and unpolarized light.**

Let  $R_{\perp}$  be the reflectance of perpendicular polarized incident light, which is the preferred plane to give greater reflectance.  $R_{//}$  be the reflectance of parallel polarized incident light, the plane for which the reflectance is least. Then according to the Fresnel law, the specular reflectance of such plane polarized light from a mirror surface is:

$$R_{\perp} = \left[ \frac{\cos i - \sqrt{(n_2/n_1)^2 - \sin^2 i}}{\cos i + \sqrt{(n_2/n_1)^2 - \sin^2 i}} \right]^2 \dots\dots\dots(21)$$

$$R_{//} = \left[ \frac{(n_2/n_1)^2 \cos i - \sqrt{(n_2/n_1)^2 - \sin^2 i}}{(n_2/n_1)^2 \cos i + \sqrt{(n_2/n_1)^2 - \sin^2 i}} \right]^2 \dots\dots\dots(22)$$

For unpolarized incident light, the reflectance intensity (R) is the average of  $R_{\perp}$  and  $R_{\parallel}$  :

$$R = \frac{R_{\perp} + R_{\parallel}}{2}$$

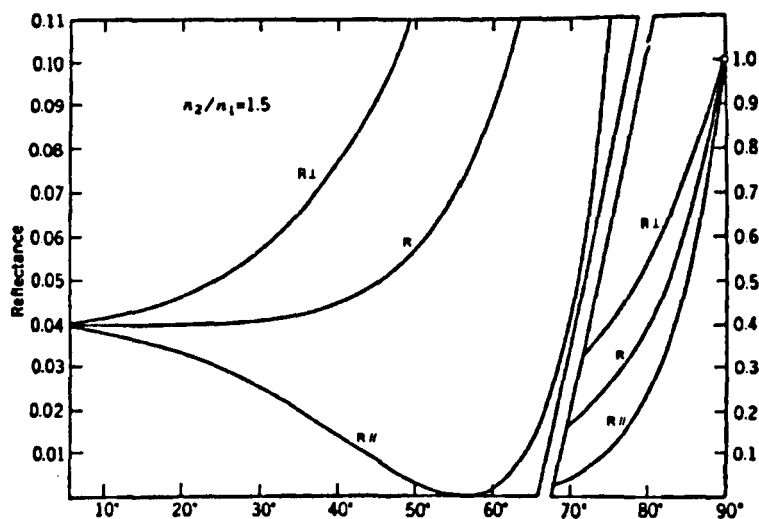
It can be seen that

$$\text{when } i = 0^{\circ}, R = \left[ \frac{n_2 - n_1}{n_2 + n_1} \right]^2 \quad \text{and when } i = 90^{\circ}, R = 1.$$

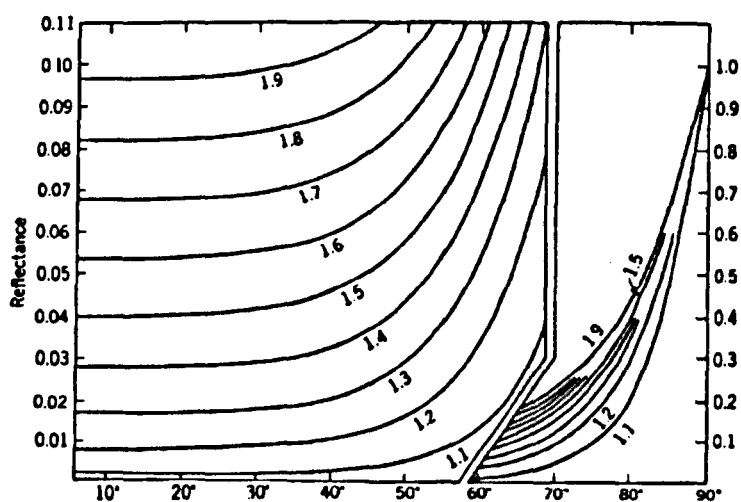
That is at both normal and glancing angles, the reflected light remains unpolarized whereas at almost all other angles, it is partially polarized as shown in Figure 2.16.

Figure 2.16 illustrates Fresnel reflectance of plane-polarized and unpolarized light from an air-glass interface ( $n_2/n_1 = 1.5$ ), as a function of angle of incidence. It is shown that the reflectance of unpolarized light increases gradually from about 4% at normal incidence to a maximum at glancing incidence. The reflectance at normal incidence and the characteristics of the curve depend on the refractive index ratio of the materials. Since  $n_1$  is usually air, the refractive index of which is approximately 1, then the higher the refractive index of the glass, the greater the reflectance. Figure 2.17 shows Fresnel reflectance for unpolarized light as a function of angle of incidence (i) for various values of refractive index ratio.

Figure 2.16 also shows that the reflectance of parallel polarized incident light passes through a minimum at an angle known as the polarizing or Brewster angle (B). Unpolarized light incident on the surface at



**Figure 2.16** Fresnel reflectance, polarized and unpolarized light, as a function of angle of incidence; for glass having a refractive index of 1.5.



**Figure 2.17** Fresnel reflectance as a function of angle of incidence for various refractive indices.

this angle will reflect a completely plane (perpendicular) polarized light. This angle which depends on the refractive index ratio is expressed as follows:

$$\tan B = n_2 / n_1 \quad \dots\dots\dots(23) \quad \text{Brewster's law}$$

As part of the incident light penetrates the surface, the intensity will decrease mainly by selective absorption and in some cases scattering may play a part. Most materials show selective absorption, that is they show strong absorption for some particular wavelengths. The intensity is decreased by a constant fraction for each unit length traversed in the material. This is expressed as Bouguer's law.

$$I = I_0 e^{-\alpha t} \quad \dots\dots\dots(24)$$

where

$I_0$  is the initial intensity, i.e., the light intensity entering the material

$I$  is the intensity after passing through a thickness  $t$

$\alpha$  is the absorption coefficient of the material, i.e., the fraction of the intensity absorbed by unit thickness

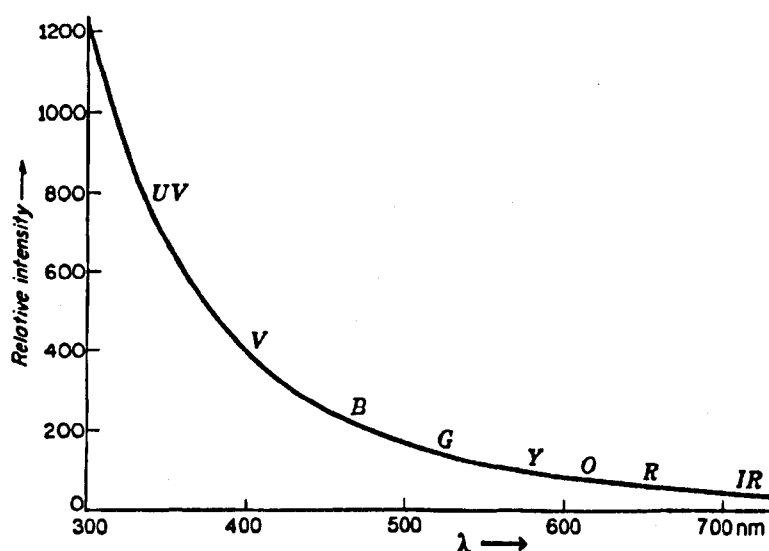
Since the loss of intensity in some cases also results from light scattering,  $\alpha$  is made up of 2 components;  $\alpha_a$  due to true absorption, i.e., the energy being transformed, and  $\alpha_s$  due to scattering. Equation 24 thus becomes

$$I = I_0 e^{-(\alpha_a + \alpha_s)t} \quad \dots\dots\dots(25)$$

2. Laws of scattering. When the particle size is much larger than the wavelength of incident light, the particle will react with light according to the Fresnel law. On the other hand, when the particle is considerably smaller than the wavelength of the incident light, it will scatter the light. The light

begins to go round the pigment particle instead of being reflected or refracted by it.

The first quantitative study of the laws of scattering by small particles was made by Rayleigh [Jenkins & White, 1957]. Rayleigh defined small particles as those which are much smaller than the wavelength of light, i.e., much less than about  $0.5\ \mu\text{m}$ . The optimum particle size for light scattering is about  $0.4\lambda$ , that is (for wavelength of the visible light) ranging from about  $0.16$  to about  $0.28\ \mu\text{m}$  [Judd & Wyszecki, 1975]. The intensity of the scattered light is proportional to  $1/\lambda^4$  where  $\lambda$  is the wavelength of light. With a given size of particles, the particles will scatter short wavelength light more effectively than long wavelength light. This is Rayleigh's law. Figure 2.18 illustrates the relationship between the intensity of the scattered light and the wavelength of light according to Rayleigh's law. It is shown from this Figure that the particles will scatter violet light more effectively than red light provided the particles are much smaller than the wavelength of either colour.



**Figure 2.18 The intensity of scattered light**

Mie theory describes the light scattering from single spherical or cylindrical particles of submicron size. For a spherical particle, Figure 2.19 shows the scattered intensity is strongest in the forward direction, around which it is centrosymmetrically distributed [Borch, 1983]. The scattering coefficient for spherical shaped particles is influenced by the size and the refractive index of the particles. Particle sizes greater than half the incident wavelength were found to result in maximum scattering [Borch & Lepoutre, 1978]. It was postulated that Mie scattering theory is not appropriate for application to paper reflectance [Hemstock, 1962], since paper particles such as fibres or fillers are large and are compacted so closely that the scattering intensity distribution from a single particle disappears. However, it was found in pigmented paint that single particles play a part in the overall scattering intensity.

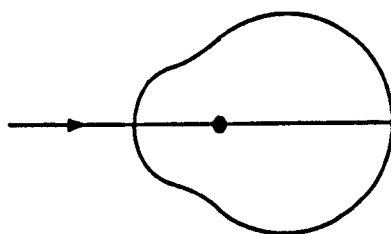


Figure 2.19 Mie scattering from submicron sphere

It was claimed that a non-contact optical method is the most effective method to characterize surfaces [Hansuebsai, 1987] and this is the method to be employed in this study. Therefore, the previous relevant work will be discussed.



#### 2.6.4 Studies of surface characteristics by reflectance measurements.

The magnitude and the angular distribution of the surface reflection from a rough surface are determined by the laws governing the behaviour of electromagnetic radiation, the complex refractive index of the surface and the roughness of the surface profile [Oittinen, 1980].

Davies [1954] was the first to derive equations relating surface reflection distribution to surface profiles of a perfectly conducting (e.g., metallic) rough surface. Surface reflection was characterized by a Gaussian depth distribution of roughness and a Gaussian autocorrelation function. Davies' statistical surface model gives two parameters; the standard deviation of the depth distribution, which is experimentally equivalent to r.m.s. roughness, ( $\sigma$ ) and the autocorrelation length ( $a$ ).

Bennett and Porteus [1961] also studied the relation between surface roughness and specular reflectance at normal incidence by using long wavelength radiation. They modified Davies equations and expressed the specular and diffused reflectance of light incident at zero degrees, on a rough metal surface, in the following equations:

$$R_s = R_o \exp \left[ - (4\pi\sigma)^2 / \lambda^2 \right] \dots\dots\dots(26)$$

$$R_d = R_o \frac{2^5 \pi^4}{m^2} (\sigma/\lambda)^4 (\Delta\theta)^2 \dots\dots\dots(27)$$

where

$R_s$  is the specular reflectance of the rough surface

$R_d$  is the diffused reflectance of the rough surface

$R_o$  is the specular reflectance of a perfectly smooth surface of the same

material

$\sigma$  is the root mean square roughness, defined as the root mean square deviation of the surface from the mean surface level

$\lambda$  is the wavelength of the incident light

$m$  is the root mean square slope of the profile of the surface

$\Delta\theta$  is the instrumental acceptance angle

and the total reflectance  $R$  is

$$R = R_0 \exp \left[ -(4\pi\sigma)^2 / \lambda^2 \right] + R_0 \frac{25\pi^4}{m^2} (\sigma/\lambda)^4 (\Delta\theta)^2 \dots\dots\dots(28)$$

These expressions are applicable only when the root mean square roughness is small compared to the wavelength of light. When the wavelength is sufficiently long, the diffused component can be neglected resulting in  $R = R_s$ . Thus the value of root mean square roughness ( $\sigma$ ) can be calculated directly from the measured reflectance. That is the surface roughness can be determined regardless of the root mean square slope of the surface profile.

Bennett [1963] and Porteus [1963] considered that specularly reflected light is coherently reflected whereas diffusely reflected light or scattered light is incoherently reflected. Bennett extended the theory to various aluminized ground-glass surfaces. The height distribution of the surface profile was Gaussian which gave close agreement with the theory previously discussed. Porteus extended the theory to measurements at shorter wavelengths and explained the characteristic behaviour as follows. When the roughness ( $\sigma$ ) is small compared to the wavelength, i.e., the difference in the levels within the facets is small relative to the wavelength, the surface will

behave as a plane reflector. The reflected light is thus coherent. As the wavelength gets shorter, the difference of the facets becomes significant so that the facets scatter the light independently resulting in incoherent reflectance until completely incoherent reflectance is obtained at the shorter wavelengths. When the wavelength is sufficiently short compared to the average dimension of the facets, the facets will behave as a plane reflector that is the incoherent component is now specularly reflected.

Warren & Peel [1973] proposed a non-contact optical method based on the relationship between specular reflectance and the root mean square roughness as expressed in the following form:

$$P = P_0 \exp [-(k\sigma \cos\theta / \lambda)^2] \dots\dots\dots(29)$$

where

$P$  and  $P_0$  are the reflected and the incident light intensities

$\theta$  is the angle of incidence

$\lambda$  is the wavelength

$k$  is a constant

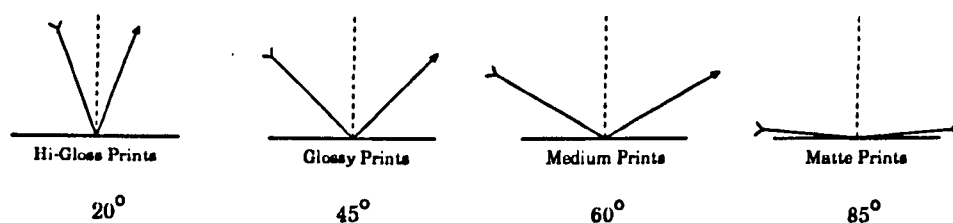
Since there is evidence that paper generally has a r.m.s. roughness range of about 1-5  $\mu\text{m}$  [Hsu, 1964], the radiation selected was infrared radiation. They employed a simple infrared reflectometer and measured the characteristics of specular reflectance to indicate the roughness of the paper. Warren & Peel found that the peak intensity of the reflectance from a rougher paper is broader and of lower intensity than that of a smoother paper. The effects of various incident angles and of wavelengths were also investigated and they showed that increased wavelengths or increased incident angles gave a narrower peak of greater reflectance. They proposed that peak reflectance at

an appropriate wavelength is the best parameter for the determination of roughness.

A quantitative study of the paper coating surfaces was done by Gate, Windle & Hine [1973]. Typical coatings were prepared and surface roughness was determined by both an optical and a mechanical profile method. The optical method involved specular reflectance measurements with a specially constructed spectro-goniophotometer. The optical roughness was calculated from the reflectance data using the Bennett-Porteus equation. They found a good correlation between the optical root mean square roughness ( $\sigma_o$ ) and the mechanical root mean square roughness ( $\sigma_p$ ). The mean value of these optical and mechanical roughnesses, is defined as  $\sigma$ . The authors [1973] showed a good correlation between  $\sigma^2$  and gloss value (plotted on a logarithmic scale). The maximum gloss value is obtained when  $\sigma$  is zero, corresponding to a completely smooth surface.

It was found that the optical print quality of solid printing arises mainly from ink and paper interactions whereas in halftone printing it is controlled by the ink properties to a greater extent [Oittinen & Lindqvist, 1982]. When considering a single-colour solid printing, one of the most important quality factors is print gloss [Schaeffer *et al.*, 1963]. Fetsko & Zettlemoyer [1962] indicated that uniformity of gloss is also important since lack of uniformity can be the greatest deterrent to any print quality characteristic. Gloss is created in the topmost surface layer of the material structure. It is defined as the degree to which a surface approaches a mirror surface. This agreed with Gate's work which showed that print gloss is dependent on the smoothness of the paper and the smoothness of the

printed film. It will be shown that studies of print gloss and print smoothness are not separated. Among six distinct types of gloss classified by Hunter [1937], specular gloss is the type most commonly referred to. Gloss is based on reflectance measurements at the specular angle. Thus, the measurement of specular reflectance is generally used as a measure of gloss. The variety of angles of specular gloss measurements is shown in Figure 2.20.



**Figure 2.20 Variety of angles of specular gloss measurements.**

Fetsko, Witherell & Poehlein [1973] investigated the relationship between specular gloss and surface roughness of thirteen paperboards and the corresponding solid prints. Specular gloss measurements were made on a Gardner Multiangle Glossmeter; surface roughness measurements with a Brush Surface Analyzer; and goniophotometer measurements on a modified Brice-Phoenix Universal Light Scattering Photometer. Specular gloss results for paperboards were consistent with the basic laws of optics that specular gloss increases with increasing angle of incidence. The prints, however, always gave lower gloss value at 85° than at 75°. In contrast, the goniophotometer reflectance results were always higher at 85° than at 75° but the maximum reflectance occurs at an angle approximately 5-8 degrees higher than the specular angle owing to the roughness of the test sample. The two angles will coincide only with perfectly smooth surfaces. The gloss of both the paperboards and the prints decreased with increasing number of

roughness elements on the test surface. They found a relationship between specular gloss and the smoothness for both the paperboards and the prints only at the glancing angle of 85°.

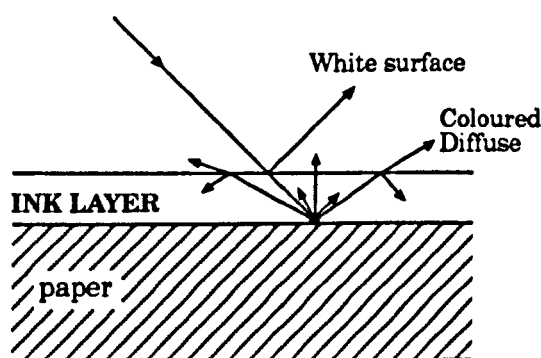
It has been shown that gloss is related to roughness ( $\sigma$ ) [Gate *et al.*, 1973; Fetsko *et al.*, 1973]. In addition, Oittinen [1980] examined two roughness parameters derived from surface reflectance measurements according to the Davies and Bennett - Porteus theory to determine the specular gloss of surfaces. Diffuse reflectance was calculated from the Lambert Law and subtracted from the total reflectance obtained on a Zeiss goniophotometer. From the results of nineteen coated and uncoated papers and boards, she found that the r.m.s roughness ( $\sigma$ ) was inadequate for the determination of specular gloss and that the size distribution of roughness had to be taken into account in order to obtain a more accurate specular gloss. She introduced two more parameters, S and g, into the Bennett-Porteus equation to account for the multiple scattering which would decrease the reflectance.

$$R = S \exp^{-(4\pi\sigma/\lambda)^2 \cos^2 i} (1+g) \dots\dots\dots(30)$$

S is inversely proportional to multiple scattering and correlated well with optical roughness ( $\sigma_0$ ). 'g' is an estimate of the proportion of scattered light which is included with the specular reflectance component.

A schematic diagram of the reflectance from an ink film is shown in Figure 2.21. There are two components of surface reflection from this ink film; a white surface component and a coloured diffuse component. It is

known that the diffuse component is included in the surface reflectance measurement. Leekly, Denzer & Tyler [1970] proposed a method using a goniophotometer to measure surface reflection and to distinguish between surface and diffuse components for papers and prints. The method is based on the assumption that polarized light incident on a surface is partly reflected from the upper surface of the ink film without depolarization and is partly refracted into the ink film. This refracted component becomes diffusely scattered and completely depolarized within the ink film. However, repolarization occurs when this completely depolarized light re-emerges from the ink film. They found that this repolarization effect cannot be neglected in order to separate specular and diffuse components accurately. For all the samples tested, separation into surface and diffused reflectance was achieved.



**Figure 2.21 A schematic diagram of reflectance from an ink film.**

The resolution of reflectance into surface and diffuse components has been made using a series of printed and unprinted papers with different surface characteristics. In all cases, the printed surfaces showed higher surface reflectance at the specular peak than the unprinted surfaces. This

was due to the filling of the surface depressions and levelling of the surface by the ink film. A mathematical surface model was developed by Leekly *et al.* to calculate from the reflectance data the fraction of the surface which is inclined at an angle  $t$  to the nominal plane of the surface.

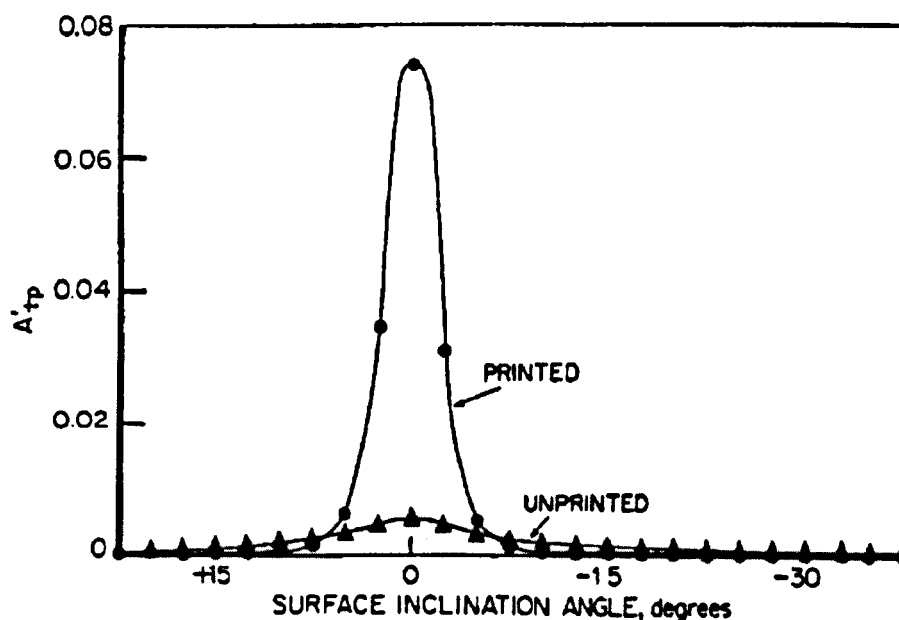
$$A'_{tp} = [R_s/R_s(\max)] [\cos t \cos 45^\circ / \cos (45^\circ + t)] \quad \dots\dots\dots(31)$$

where

$A'_{tp}$  is the fraction of the surface, measured by projection on the plane of the surface, which is inclined at an angle  $t$  to the nominal plane of the surface

$R_s$  is the measured specular reflectance at detector angle,  $r'$

$R_s(\max)$  is the specular reflectance which would occur if the whole surface were optically flat and inclined at the angle  $t$  and can be calculated from the Fresnel equation

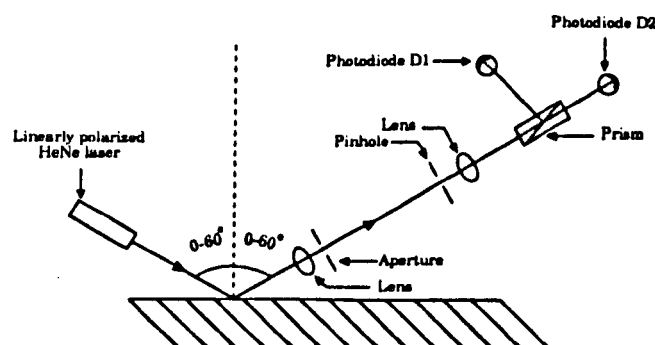


**Figure 2.22 An illustration shows an increase in optical smoothness when a dull coated paper was printed.**



The surface inclination or optical smoothness is expressed as a plot of  $A't_p$  against surface inclination angles. Figure 2.22 illustrates an increase in optical smoothness when a dull coated paper was printed. The fraction of printed surface which lies in contact to the nominal plane of the surface (inclination angle is zero) is greater than that of the unprinted surface.

Consistent with the concept of reflectance measurement of Leekly *et al.* [1970], Bryntse and Norman [1976] developed a new instrument which can measure simultaneously both the surface reflectance and the diffuse reflectance of a surface. This makes it possible to separate these two components. The important part of the instrument is an optical system as shown in Figure 2.23. The scanning device of the instrument makes it possible to measure the variations in surface and diffuse reflectance of printed and unprinted paper samples. Experimental results showed that the average surface reflectance (including very small scattering reflectance) of a calendered print is approximately twice as high as that of uncalendered print whereas the average diffuse reflectance is not influenced by calendering.



**Figure 2.23 An optical system of Bryntse & Norman.**

Maley [1990] has used print evaluation forms from which he has shown that ink film smoothness and ink film uniformity are the major contributors to print quality.

It appears, however, that little work has been done to evaluate print smoothness. Therefore, an area of particular interest throughout the course of the present study is print smoothness. The most obvious distinction between an ink film and a paper is that the former is not compressible whereas the latter is. As a result, most of the methods used in measuring paper surfaces as described above are not appropriate to the examination of an ink film except for the reflectance methods. For example, regarding print gloss measurement, there was evidence that the optimum geometrical conditions varied for prints made with different ink-paper combinations. Gloss values increased with increasing incident and measured angles. This implies that until a standard geometrical condition is established, comparisons of print gloss based on gloss values may not be reliable [Fetsko *et al.*, 1962; Fetsko *et al.*, 1973].

#### **2.6.5 The non-contact optical method. ( used in this study)**

A non-contact optical method was recently developed by Hansuebsai [1989]. He developed a statistical surface model to calculate macrosmoothness and microsmoothness from reflectance measurements for paper surfaces. This method will be applied in the present work to evaluate printed surfaces; it has the following advantages.

1. The method considers the whole surface that is above and below the mean surface. The mean surface is a geometrical plane which is an average within the profile variation.

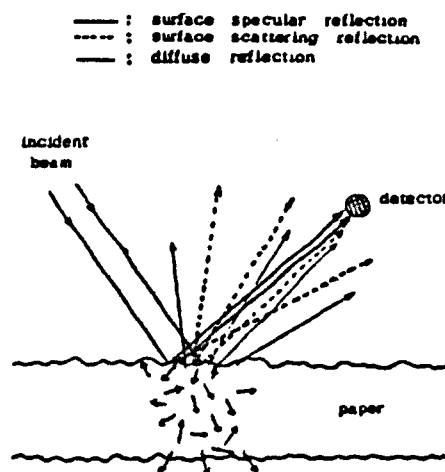
2. The method can separate (primary) surface reflection from (internal) diffuse reflection by means of polarized light.

3. The printed surfaces under measurement are in an uncompressed condition as usually seen by the customers.

Developments of the method will be described from two aspects: concept and surface characteristics.

*Concept.* The method is based on the understanding of surface reflection and Barkas' classical model.

1. Surface reflection. In general, when a surface is illuminated, the reflection from the surface can be surface specular reflection, surface scattering reflection and re-emerging diffuse reflection as demonstrated in Figure 2.24 with a paper surface.



**Figure 2.24 Surface reflection of a paper surface.**

2. Barkas' classical model. Barkas [1939] considered that a surface contains two types of small elementary facets of suitable areas which may be inclined at varying angles to the mean surface. One type consists of mirror facets which reflect a proportion of incident light according to Fresnel's law and the other type consists of rough facets which diffuse a proportion of incident light according to Lambert's law. Barkas' surface model was applied to a paper surface. He considered that complete fibres have reflecting surfaces whereas finely divided fibres and the filling material scatter the light. In the case when a highly inclined mirror facet is illuminated, the reflected light will fall onto other facets, either mirror or rough, and so on. Consequently such light, on emerging, will be classified as coming from rough facets. Light which is refracted through the surface undergoes many multiple internal reflections. Eventually when re-emerging, it is equivalent to having undergone reflection from rough facets.

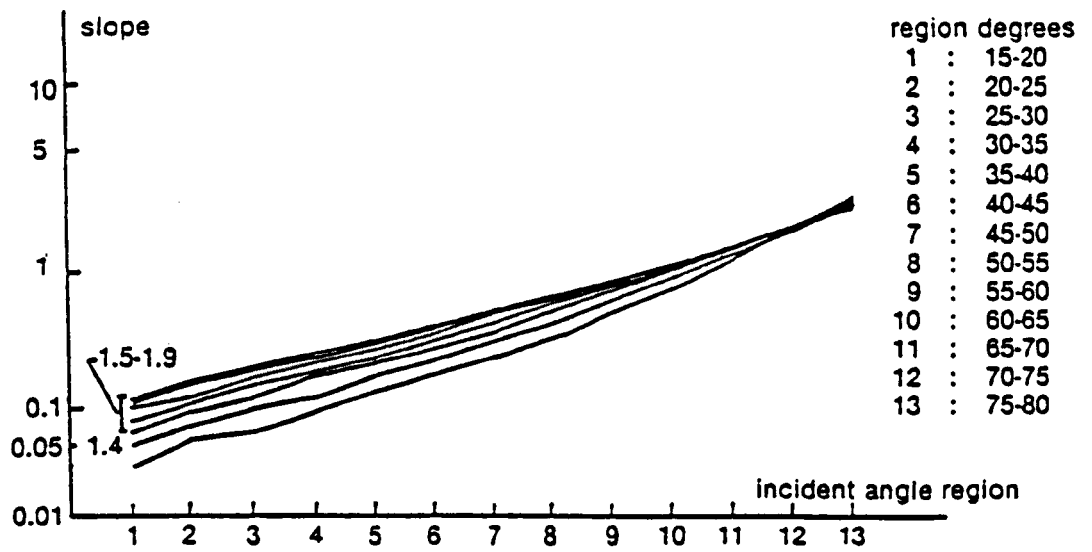
It is generally known that there is no scattered light from a perfect plane surface (e.g., mirror surface) and the light, accordingly, is only specularly reflected from it. The method is thus designed to relate to the smoothness of the surface with respect to the light which is reflected from it in the specular direction.

#### *Surface characteristics.*

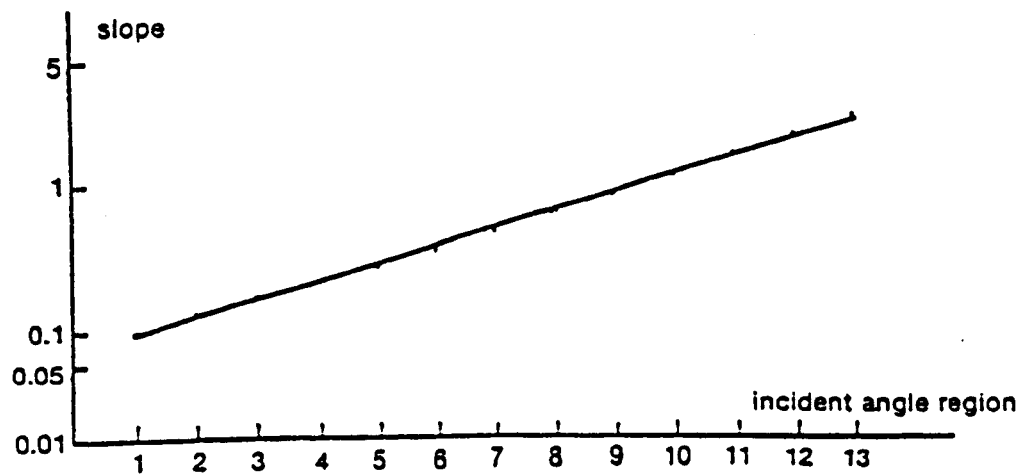
Surface characteristics are characterized into macrosmoothness and microsmoothness. Macrosmoothness mostly arises from the real reflecting elements (mirror facets) above the mean plane. Microsmoothness arises from a combination of macro elements and micro elements below the mean plane. This combination scatters the light which can be considered as equivalent to

that from the mirror facets above the mean plane. Both macrosmoothness and microsmoothness are determined only from the surface reflection measured at the specular angle. However, diffuse light which arises from highly inclined reflecting elements is included in each measurement. In order to discriminate between the surface reflection and the diffuse reflection, the method is based on the reflection characteristics of polarized light. That is the surface reflection remains polarized whereas the diffuse reflection would be depolarized as a result of multiple internal reflections within the materials. This depolarized light is eliminated by means of the method of measurement which will be described later in the Preliminary Study.

1. Derivative reflectance technique. This is a useful technique to eliminate the influence of the refractive index of the surface on the evaluation of smoothness parameters. With reference to Figure 2.17, it is shown from Fresnel's law for specular reflectance that the reflectance intensity is dependent on the refractive index of the surface. This reflectance information does not seem to be useful because of the effect of the refractive index. To search for an appropriate standard surface for the determination of optical smoothness, it was found that derivative or slope values of the reflectance provide a potentially useful form. The slopes of the surface reflectance at specular angles for refractive index values from 1.5 to 1.9 are determined over a series of incident angle regions from 15 degrees to 80 degrees at 5-degree interval. The plot of these slope values, versus the angles of a series of incident angle regions on a normal printability graph, is shown in Figure 2.25. It can be noted that a linear line is obtained for each refractive index chosen and these lie close to one another. The averaged slope values for these refractive index values are calculated and plotted as shown



**Figure 2.25 A plot of slope vs angle of incidence.**



**Figure 2.26 Average slope values for the refractive indices of 1.5-1.9.**

in Figure 2.26. These averaged slope values are postulated to be used as a smooth standard surface for both paper and printed surfaces. It has been reported that the refractive index of paper and ink film are nearly the same [Yule, 1967]. The refractive index of paper is 1.55 [Barkas, 1939]; that of an oil-based vehicle is approximately 1.45-1.65 [Judd & Wyszecki, 1976; Young, 1973].

## 2. Statistical surface model - Macrosmoothness and Microsmoothness.

Measurements within a range of incident angles from 15 to 80 degrees, of a small surface sample are sufficient to evaluate the whole profile of that surface; both above and below the mean surface. It can be envisaged that as the incident angle increases towards glancing angle, the specularly reflected light arises from elements only at or above the mean surface. Therefore, the method is designed to divide the observed conditions into two angular ranges to separate the surface region above the mean surface from that below the mean surface. One set of measurements ranging from 15 to 45 degrees is used to obtain the reflectance from a combination of macro and micro elements below the mean surface. The other set ranging from 45 to 80 degrees is used to obtain reflectance from, mostly, macro elements above the mean surface. Both sets of angles are determined with respect to zero degrees at the normal to the surface.

Smoothness is defined as the extent to which the surface consists of plane-mirror facets as measured per unit of illuminated area. This implies that the relationship between the slope values of the sample and those of the standard surface can be attributed to surface smoothness. The techniques used to study the relationship are correlation and regression analysis.

Let  $D_p(n)$  and  $D_s(n)$  be the derivative or slope values of the reflectance determined in each of a series of 5-degree intervals of the sample and of the standard surface, respectively. 'n' is the index for angular regions numbered from 1 to 13 where  $n = 1$  for the set at 15-20 degree to  $n = 13$  for the set at 75-80 degree. Thus the regions in which  $n \leq 6$  are related to microsmoothness. The regions in which  $n \geq 7$  are related to macrosmoothness.

Microsmoothness ( $S_u$ ). Correlation is a widely used measure of the closeness of a relationship. It was found that the correlation coefficient, 'r value', between the slope values of the sample and of the standard surface is an appropriate measure of microsmoothness; since  $D_p(n)$  within the range where  $n \leq 6$ , of one sample is not significantly different from others. It is not useful to analyse further by regression. Thus, microsmoothness ( $S_u$ ) can be written as follows:

$$S_u = k \frac{C(D_p \times D_s)}{S(D_p) \times S(D_s)} \dots\dots\dots(20)$$

where

$C(D_p \times D_s)$  is the covariance of  $D_p$  and  $D_s$ .

$S(D_p)$  is the standard deviation of  $D_p$ .

$S(D_s)$  is the standard deviation of  $D_s$ .

$k$  is the instrument constant. (in this practical work,  $k = 1$ )

Macrosmoothness ( $S_m$ ). Similarly, it is shown from the correlation coefficient that there is a significant relationship between the sample and the standard surface within the range where  $n \geq 7$ . A mathematical model based



on linear regression is applied. Therefore, macrosmoothness ( $S_m$ ) can be written in the following form:

$$S_m = k \frac{C(D_p \times D_s)}{V(D_s)} \dots\dots\dots(21)$$

where

$V(D_s)$  is the variance of  $D_s$ .

Both  $S_m$  and  $S_u$  provide information about the surface. The higher the value, the smoother the surface.

### **3. PRELIMINARY STUDY**

#### **Study of the effects of ink vehicle penetration on optical smoothness of paper.**

Earlier studies [Hansuebsai & Morantz, 1988; Hansuebsai, 1989] provided methods for the optical assessment of the smoothness of paper and proposed that the optical smoothness of a paper can be characterized by means of two parameters, namely macrosmoothness ( $S_m$ ) and microsmoothness ( $S_u$ ). In his study, Hansuebsai [1989] has reported the effects of certain solvents on the optical smoothness of paper. It is intended to use this technique to study ink and paper interactions. The extension of these studies to a printed surface was envisaged as the next step. However, this preliminary study makes a first approach to investigate the effects, on the optical smoothness of paper, of individual ink vehicles and of one combination of these offset litho ink vehicles using materials employed in the printing industry. The results establish that a more comprehensive investigation on inked surface is warranted and certain considerations need to be taken into account.

### **3.1 NON-CONTACT OPTICAL SMOOTHNESS METHOD.**

#### **3.1.1 The instrument.**

The method employs a specially constructed instrument which is a form of goniophotometer, an instrument that measures reflectance as a function of incident angle and viewing angle, as shown in Figure 3.1. It is composed of three parts:

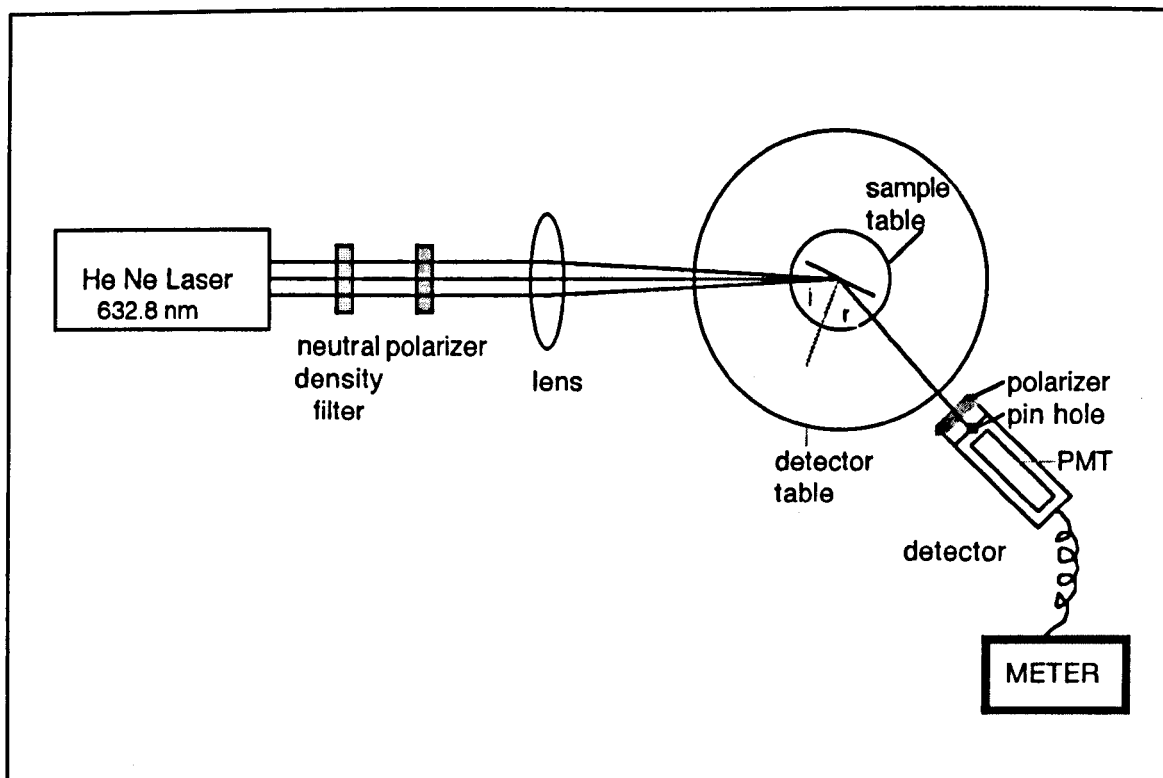


Figure 3.1 The optical reflectance instrument.

*Illumination system.* A 5 mW 632.8 nm HeNe linearly polarized laser, type GLT 2140 from NEC, is used as the light source and a power supply from Power Technology Inc. is used to operate the laser. A linear dichroic polarizer is placed in front of the laser in order to ensure the polarisation state of the incident beam. Neutral density filters are used to reduce the intensity of the beam so that the intensity of the reflected light corresponds to the capability of the meter. A 15 cm focal length convex lens is employed to converge the beam to a smaller area focused onto the illumination regions.

*Mechanical stage.* This part is designed in order to be able to move the normal axes of the paper sample and the detector plane simultaneously parallel to a horizontal plane. This enables changes in the incident and the measured angles to be made.

*Detection system.* The system uses a photomultiplier (PMT), which is an extremely sensitive light detector providing a current output proportional to light intensity, type 9524B manufactured by Thorn EMI Electron Tubes Ltd, (See circuit in Appendix A) as the detector. A power supply from Brandenburg type 2479N is used to operate the photomultiplier. Since the intensity of the reflected light is very low resulting in a large photon noise, the photomultiplier output is thus passed through a low pass filter (See circuit in Appendix A) to reduce this photon noise and is linked through a linear amplifier to a voltmeter to produce a reflectance readout (See circuit in Appendix A). The detector incorporates a pinhole of 1 mm diameter to form the aperture of the detector with another linear polarizer placed in front of the detector in order that the plane of polarization can be chosen.

*Calibration of the instrument.* This study uses a sheet of glass with a back surface ground and painted with a matt black paint as the reference standard smooth surface. The instrument is calibrated by adjusting the reflectance ( $R_{\perp\perp}$ ) readout at the meter to be 74% at 85 degree incident angle (glancing angle) and 0% when there is no incident light according to Fresnel's reflectance at refractive index 1.55 (See Appendix B).

### **3.1.2 Optical smoothness procedure.**

*Reflectance measurement.* Reflectances of each sample are measured at specular angles in a range from 15 to 80 degrees at 5 degree intervals. By selecting the plane of the polarizer, the incident light is perpendicular polarised light.

1. Since the instrument uses a laser light source, it is recommended to switch the laser on for at least 15 minutes before operating measurements. After this warm-up period, the standard glass surface is placed in the sample holder and the calibration is made as previously described. The standard is then replaced by the sample.

2. Measurement of reflectance  $R_{\perp\perp}$ . This includes the first surface reflected, scattered and diffuse light components whose polarisation plane is in the same plane as the incident beam.

3. Measurement of the diffuse light component  $R_{\perp//}$  whose polarisation plane is different from that of the incident beam.

4. Some of the diffuse component when re-emerging from the surface may alter its polarisation plane again. It is assumed that the measured  $R_{\perp//}$  is equal to the diffuse component in  $R_{\perp\perp}$ . Consequently, the surface and scattered components which retain polarisation are determined by  $R_{\perp\perp} - R_{\perp//}$ .

Fourteen reflectance readings are obtained over the range of angles for each point on a sample specimen.

*Evaluations of optical smoothness.* The reflectance results were calculated according to the formulas previously given in section 2.6.5, to give macrosmoothness ( $S_m$ ) and microsmoothness ( $S_u$ ).

$$S_m = k \frac{C(D_p \times D_s)}{V(D_s)}$$

$$S_u = k \frac{C(D_p \times D_s)}{S(D_p) \times S(D_s)}$$

where

$C(D_p \times D_s)$  is the covariance of  $D_p$  and  $D_s$ .

$S(D_p)$  is the standard deviation of  $D_p$ .

$S(D_s)$  is the standard deviation of  $D_s$ .

$V(D_s)$  is the variance of  $D_s$ .

$k$  is the instrument constant. (in this practical work,  $k = 1$ )

## 3.2 MATERIALS.

### 3.2.1 Paper samples.

Four different types of printing paper, namely newsprint (NP), machine finish (MF), machine glazed (MG) and poladin cartridge (PC) were chosen for study.

*Newsprint (NP).* Cheapness is very important, but permanence is not necessary, for newsprint. It is usually made from mechanical wood pulp with

sufficient chemical wood pulp to strengthen the sheet for high speed printing. It is not normally sized but may contain a small amount of loadings (5-10%) to reduce show through. The main requirement is its high oil absorbency to give quick drying by the absorption of news ink on a high speed web machine. It is supercalendered to make the sheet capable of reproducing coarse halftone illustrations.

*Machine finish (MF).* The surface finish of this paper contains a higher proportion of chemical wood pulp than is present in newsprint. It is described as a semismooth paper due to the finish and degree of calendering. It is suitable for book printing which consists mainly of texts and line illustrations.

*Machine glazed (MG).* This printing paper is usually well sized with a little loading. The manufacture of this paper is unique in that the paper web is pressed up against a finely polished M.G. cylinder, sometimes referred to as a 'Yankee' dryer, which is steam-heated. Because of its large diameter, one side of the paper which is in contact with this M.G. cylinder surface for a long time gains a high finish or glaze while the other side remains rough. As a result, machine glazed is widely used for printing posters because the glazed side makes it suitable for halftone colour printing and the rough side for carrying adhesives.

*Poladin cartridge (PC).* This is a matt coated paper. The coating is applied using a blade coating technique. After coating, the paper is pressed through a one-to-three nip calender stack on the paper machine in order to smoothen the coating.

### **3.2.2 Ink 'vehicles'.**

Three different individual ink vehicle components and one combination of ink vehicle components were used. These were supplied by Coates Lorilleux International.

*R1066.* This is the code for a high boiling point petroleum distillate normally used in sheetfed offset formulations.

*R1976.* This is the code for a process oil used in coldset black formulations for web offset printing of newspaper. This vehicle was studied only in conjunction with newsprint. The process oil was not suitable for the other paper samples since it yielded undried samples within the limited drying time.

*R1835.* This is the code for a high boiling point petroleum distillate used in heatset web offset formulations.

*SM2007.* This is the code for a 55% resin solid solution of a simple hydrocarbon resin mixed in R1066. It is used as an additive varnish in web offset news inks.



### **3.3 EXPERIMENTAL METHOD.**

**3.3.1 Flotation.** This method was used to study some of the individual ink vehicle components. Paper samples were cut into squares measuring approximately  $3 \times 3 \text{ cm}^2$ . Each sample was floated in a bath of ink 'vehicle' for 5 seconds, removed and dried at room temperature.

**3.3.2 IGT printability tester.** The resin solution (SM2007) is too viscous to apply to the paper sample by floatation; therefore an IGT printability tester model A2-3 (Pendulum Drive), using a 2 cm metal inking disc and a pressure setting of 30 kgf, was employed to apply the resin solution. The samples were dried at room temperature.

**3.3.3 Optical smoothness determinations.** The paper samples were measured to determine their smoothness both before they were treated with the ink 'vehicles' and after being dried for at least 24 hours. Nine points were measured from each sample and the results are given in Appendix C.

**3.3.4 Data analysis.** To investigate whether there were significant effects from the treatments, the macrosmoothness and microsmoothness results for each type of paper (both untreated and treated) were subjected to statistical methods namely analysis of variance. This is one of the statistical procedures commonly used to test whether or not several sample group means are equal, i.e., to test whether there are true differences between different samples or the observed difference can be attributed to chance.

For example, to test whether

$$Sm_{NP} = Sm_{NP+R1066} = Sm_{NP+R1976} = Sm_{NP+R1835} = Sm_{NP+SM2007}$$

the F ratio is calculated:

$$F = \text{Between groups mean square} / \text{Within group mean square}$$

The calculated F ratio measures the importance of differences between groups as compared to differences within groups. This calculated F ratio is compared to the F distribution value with k-1, N-k degrees of freedom and at 0.05 significance level which is the critical value of F. The between-groups degrees of freedom are k-1, where k is the number of groups and the within-groups degrees of freedom are N-k, where N is the number of cases in the entire sample. A significant difference between the sample group means is obtained when the calculated F ratio is equal to or greater than the critical value of F.

In addition, to determine which sample group means are different from each other, Duncan's multiple comparison test is used. The data analysis was performed on a 386-class personal computer using SPSS/PC+ programme and the results are shown in Appendix D.

### 3.4 RESULTS.

This preliminary study employs the same types of paper, namely newsprint (NP), machine finish (MF), machine glazed (MG) and poladin cartridge (PC) as used by Hansuebsai (1989). However, the samples derive from different paper batches.

**Table 3.0 Comparison of optical smoothness results.**

<b>Sample</b>	<b>Hansuebsai</b>		<b>This study</b>	
	<b>Sm</b>	<b>Su</b>	<b>Sm</b>	<b>Su</b>
NP	0.02	0.66	0.011	0.73
MF	0.011	0.077	0.009	0.21
MG	0.074	0.7	0.076	0.9
PC	0.017	0.14	0.010	0.17

It was found that the optical smoothness results, Table 3.0, are fairly close in all cases but that for microsmoothness of machine finish. In the present work (See Table 3.2), microsmoothness for machine finish shows a minimum value of 0.16 and the 95% confidence interval minimum is 0.184. These results are for 9 random samples. Hansuebsai's microsmoothness for machine finish is an average of a machine direction value of 0.045 and a cross machine direction value of 0.13. This latter figure can be compared with the lowest individual value obtained in this work, namely 0.16. In this light the discrepancy is not too significant as the samples were, in any event, not from the same batch. In general, therefore, the results in this study are compatible with Hansuebsai's. Tables 3.1-3.4 summarize macrosmoothness and microsmoothness of each paper sample both before and after being treated with ink 'vehicles' and also indicate the ink 'vehicles' which affect the smoothness. The differences in macrosmoothness and microsmoothness are demonstrated in Figures 3.2-3.5.

Table 3.1 Optical smoothness summary: newsprint before and after being treated with ink 'vehicles'. Each result given is the average value of nine different points on the sample (See Appendix C).

(a) Macrosmoothness (Sm).

<b>Sample</b>	<b>Sm (<math>\bar{x}</math>)</b>	<b>S.D.</b>	<b>95% C.I. for <math>\bar{x}</math></b>	<b>Min.</b>	<b>Max.</b>
Untreated NP	0.011	0.002	0.009 to 0.013	0.008	0.015
NP+R1066 <sup>a</sup>	0.018	0.003	0.016 to 0.021	0.014	0.023
NP+R1835 <sup>a</sup>	0.021	0.003	0.018 to 0.023	0.015	0.024
NP+R1976 <sup>a</sup>	0.018	0.004	0.015 to 0.021	0.013	0.024
NP+SM2007	0.013	0.004	0.009 to 0.016	0.008	0.020

a - Sm significantly increased when compared with untreated NP and NP treated with SM2007 (See Appendix D and Figure 3.2).

(b) Microsmoothness (Su) .

<b>Sample</b>	<b>Su (<math>\bar{x}</math>)</b>	<b>S.D.</b>	<b>95% C.I. for <math>\bar{x}</math></b>	<b>Min.</b>	<b>Max.</b>
Untreated NP	0.728	0.069	0.675 to 0.781	0.619	0.832
NP+R1066 <sup>a</sup>	0.818	0.056	0.775 to 0.861	0.750	0.898
NP+R1835	0.744	0.043	0.711 to 0.777	0.688	0.809
NP+R1976	0.732	0.055	0.689 to 0.774	0.649	0.819
NP+SM2007	0.799	0.126	0.702 to 0.895	0.613	0.939

a - Su significantly increased when compared with untreated NP and NP treated with R1976 (See Appendix D and Figure 3.2).

Table 3.2 Optical smoothness summary: machine finish before and after being treated with ink 'vehicles'. Each result given is the average value of nine different points on the sample (See Appendix C).

(a) Macrosmoothness (Sm).

Sample	Sm ( $\bar{X}$ )	S.D.	95% C.I. for $\bar{X}$	Min.	Max.
Untreated MF	0.009	0.002	0.007 to 0.010	0.006	0.012
MF+R1066 <sup>a</sup>	0.015	0.005	0.011 to 0.019	0.009	0.021
MF+R1835	0.008	0.004	0.005 to 0.011	0.005	0.017
MF+SM2007 <sup>a,b</sup>	0.020	0.003	0.018 to 0.023	0.017	0.027

a - Sm significantly increased when compared with untreated MF and MF treated with R1835 (See Appendix D and Figure 3.3).

b - Sm significantly increased when compared with MF treated with R1066 (See Appendix D and Figure 3.3).

(b) Microsmoothness (Su).

Sample	Su ( $\bar{X}$ )	S.D.	95% C.I. for $\bar{X}$	Min.	Max.
Untreated MF	0.211	0.034	0.184 to 0.237	0.162	0.265
MF+R1066 <sup>a</sup>	0.739	0.067	0.688 to 0.791	0.629	0.812
MF+R1835 <sup>b</sup>	0.403	0.098	0.327 to 0.478	0.197	0.534
MF+SM2007 <sup>a</sup>	0.721	0.072	0.666 to 0.777	0.604	0.802

a - Su significantly increased when compared with untreated MF and MF treated with R1835. (See Appendix D and Figure 3.3)

b - Su significantly increased when compared with untreated MF (See Appendix D and Figure 3.3).

Table 3.3 Optical smoothness summary: machine glazed before and after being treated with ink 'vehicles'. Each result given is the average value of nine different points on the sample (See Appendix C).

(a) Macrosmoothness (Sm).

Sample	Sm ( $\bar{X}$ )	S.D.	95% C.I. for $\bar{X}$	Min.	Max.
Untreated MG	0.076	0.013	0.066 to 0.086	0.067	0.108
MG+R1066 <sup>a</sup>	0.042	0.009	0.035 to 0.049	0.025	0.053
MG+SM2007 <sup>a</sup>	0.052	0.020	0.037 to 0.068	0.037	0.099

a - Sm significantly decreased when compared with untreated MG (See Appendix D and Figure 3.4).

(b) Microsmoothness (Su).

Sample	Su ( $\bar{X}$ )	S.D.	95% C.I. for $\bar{X}$	Min.	Max.
Untreated MG	0.897	0.062	0.849 to 0.945	0.766	0.949
MG+R1066	0.874	0.074	0.817 to 0.930	0.764	0.970
MG+SM2007 <sup>a</sup>	0.802	0.052	0.762 to 0.842	0.727	0.880

a - Su significantly decreased when compared with untreated MG and MG treated with R1066 (See Appendix D and Figure 3.4).

Table 3.4 Optical smoothness summary: poladin cartridge before and after being treated with ink 'vehicles'. Each result given is the average value of nine different points on the sample (See Appendix C).

(a) Macrosmoothness (Sm).

<b>Sample</b>	<b>Sm (<math>\bar{x}</math>)</b>	<b>S.D.</b>	<b>95% C.I. for <math>\bar{x}</math></b>	<b>Min.</b>	<b>Max.</b>
Untreated PC	0.010	0.002	0.008 to 0.011	0.007	0.013
PC+R1066 <sup>a</sup>	0.018	0.004	0.015 to 0.022	0.009	0.023
PC+SM2007 <sup>a,b</sup>	0.049	0.010	0.041 to 0.057	0.031	0.065

a - Sm significantly increased when compared with untreated PC (See Appendix D and Figure 3.5)

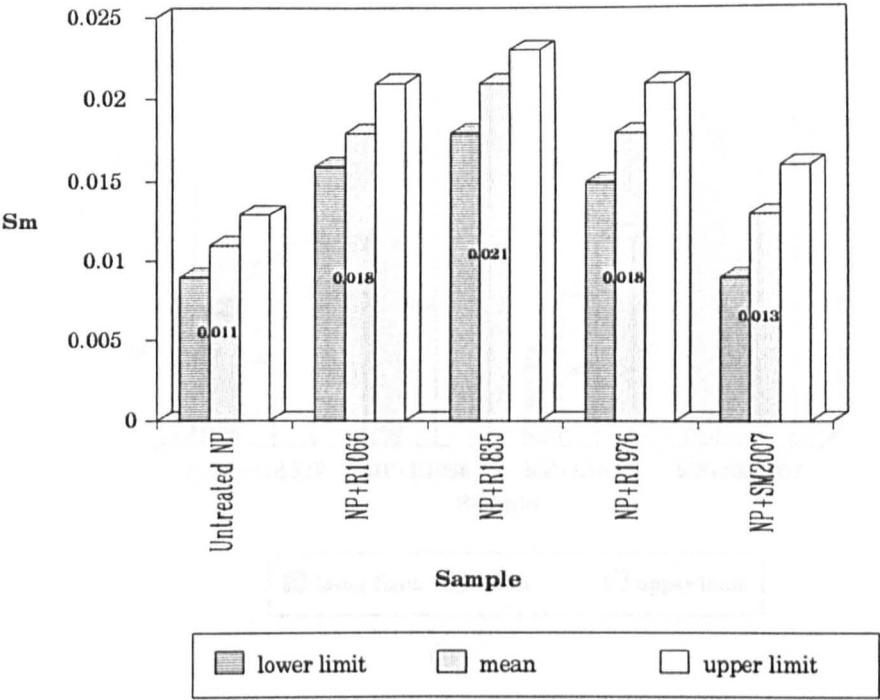
b - Sm significantly increased when compared with PC treated with R1066 (See Appendix D and Figure 3.5)

(b) Microsmoothness (Su).

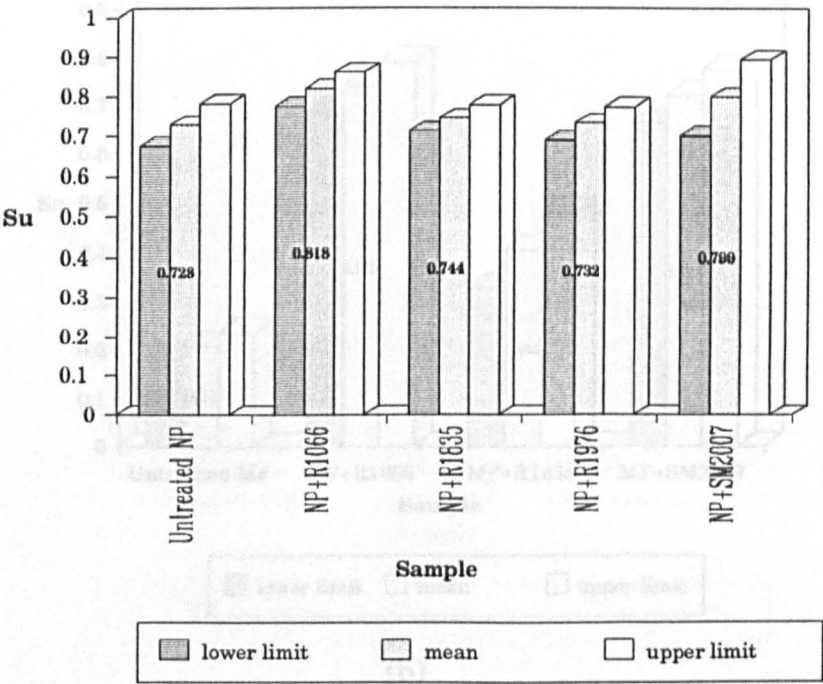
<b>Sample</b>	<b>Su (<math>\bar{x}</math>)</b>	<b>S.D.</b>	<b>95% C.I. for <math>\bar{x}</math></b>	<b>Min.</b>	<b>Max.</b>
Untreated PC	0.165	0.028	0.143 to 0.186	0.135	0.212
PC+R1066 <sup>a</sup>	0.793	0.110	0.708 to 0.877	0.606	0.932
PC+SM2007 <sup>a</sup>	0.823	0.081	0.760 to 0.886	0.665	0.930

a - Su significantly increased when compared with untreated PC (See Appendix D and Figure 3.5).

Figure 3.2 (a) Macrosmoothness (b) Microsmoothness of newsprint before and after being treated with 'ink vehicles'.



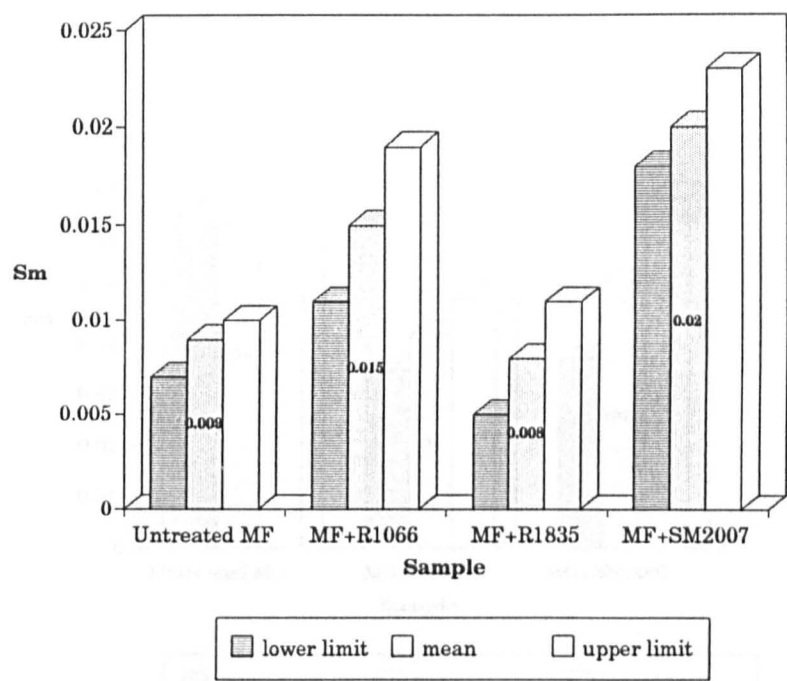
(a)



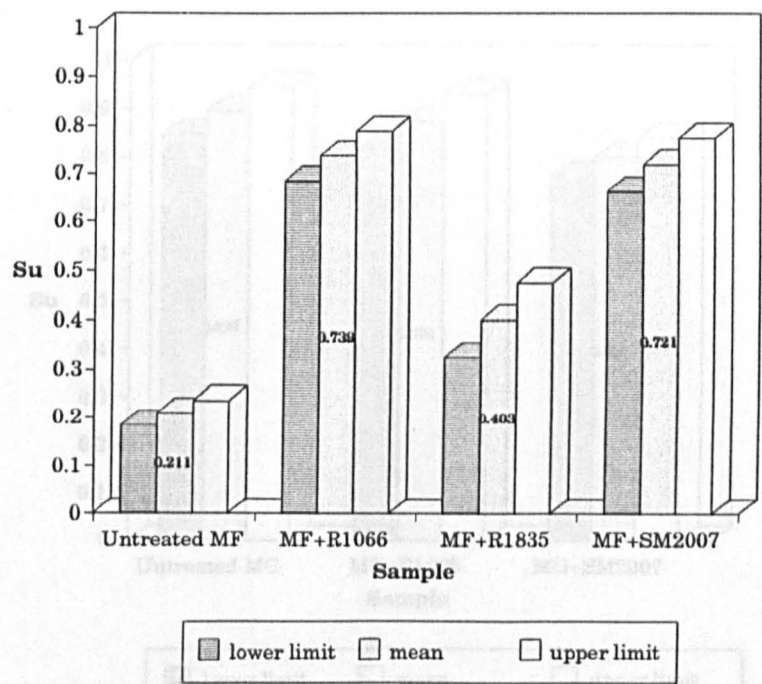
(b)



Figure 3.3 (a) Macrosmoothness (b) Microsmoothness of machine finish before and after being treated with 'ink vehicles'.

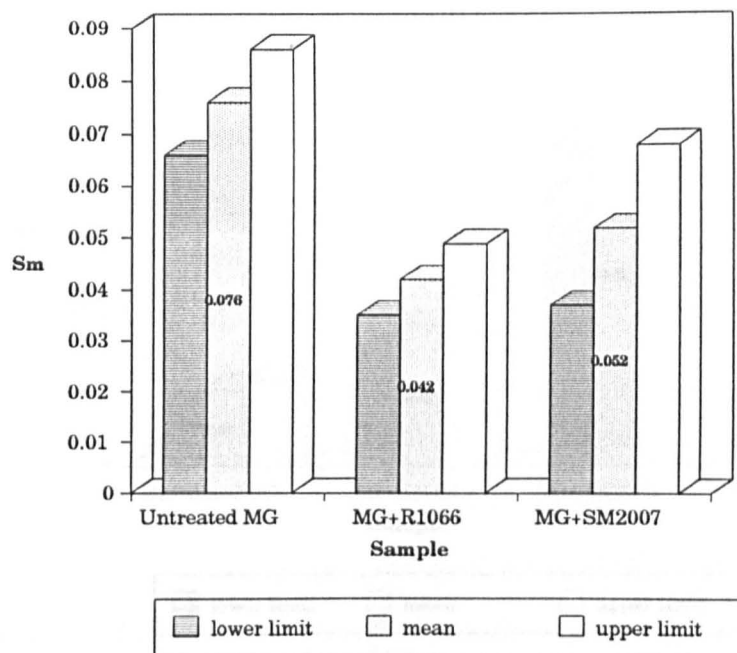


(a)

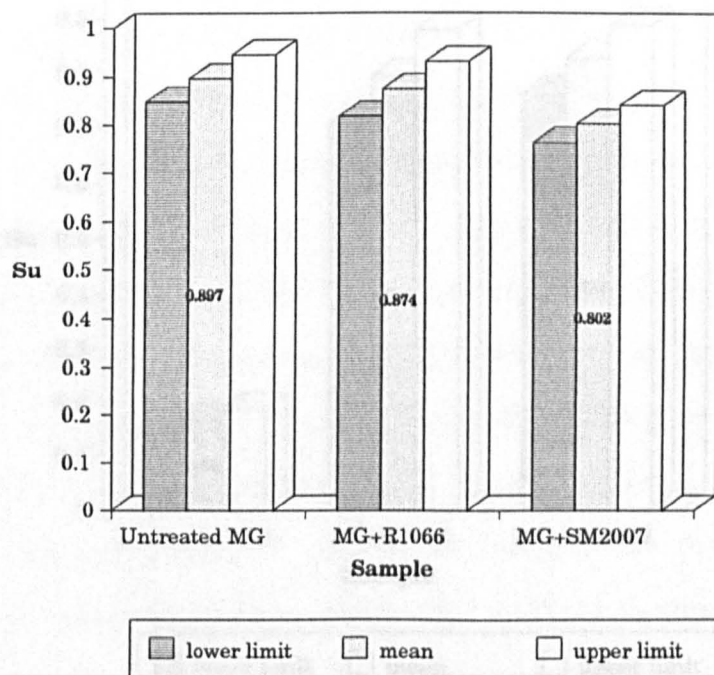


(b)

Figure 3.4 (a) Macrosmoothness (b) Microsmoothness of machine glazed before and after being treated with 'ink vehicles'.

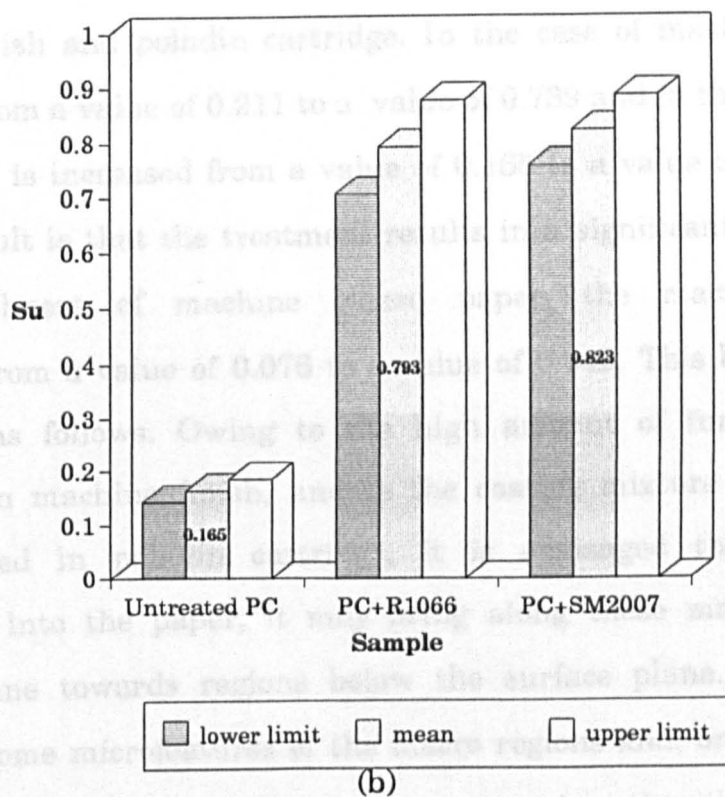
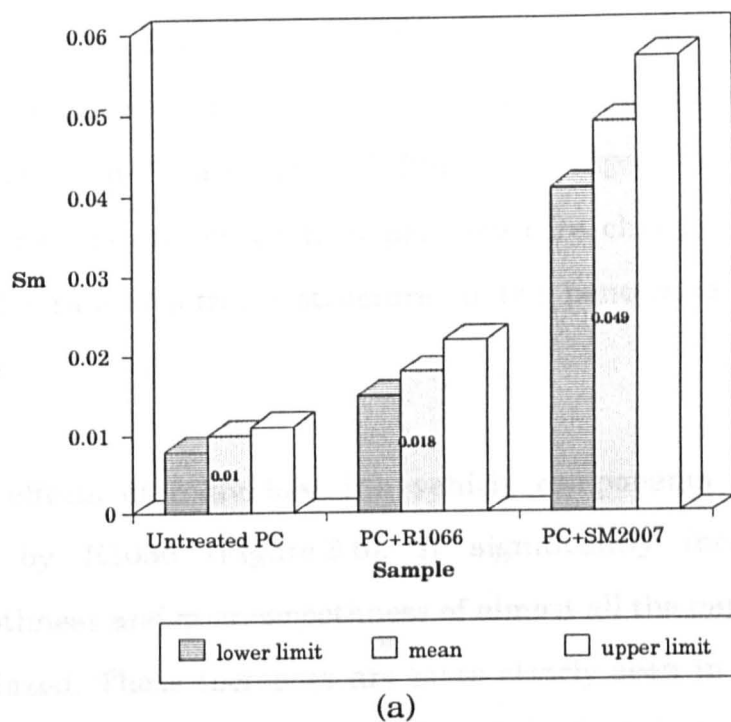


(a)



(b)

Figure 3.5 (a) Macrosmoothness (b) Microsmoothness of poladin cartridge before and after being treated with 'ink vehicles'.

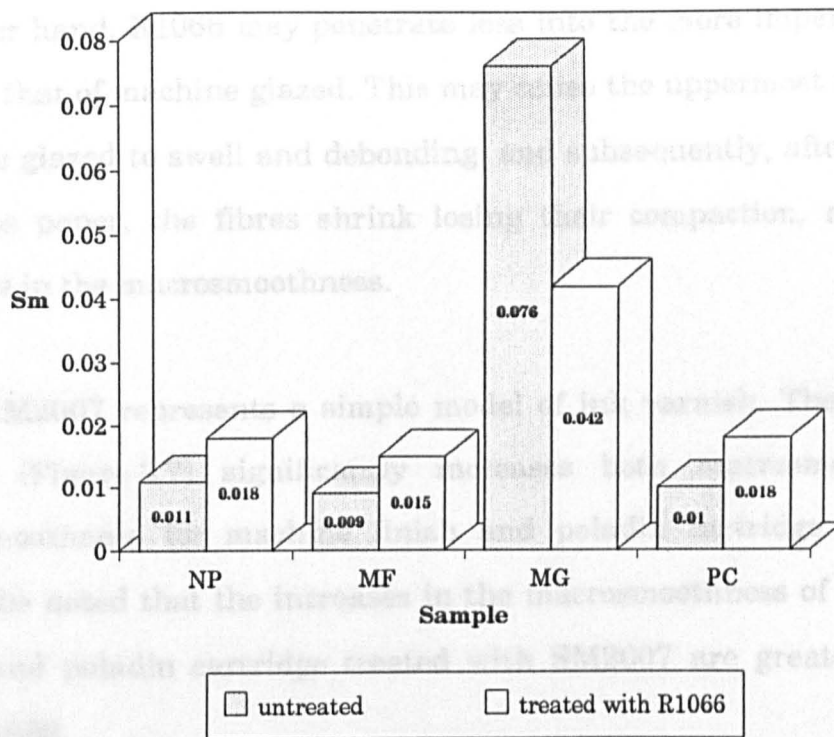


### 3.5 DISCUSSION.

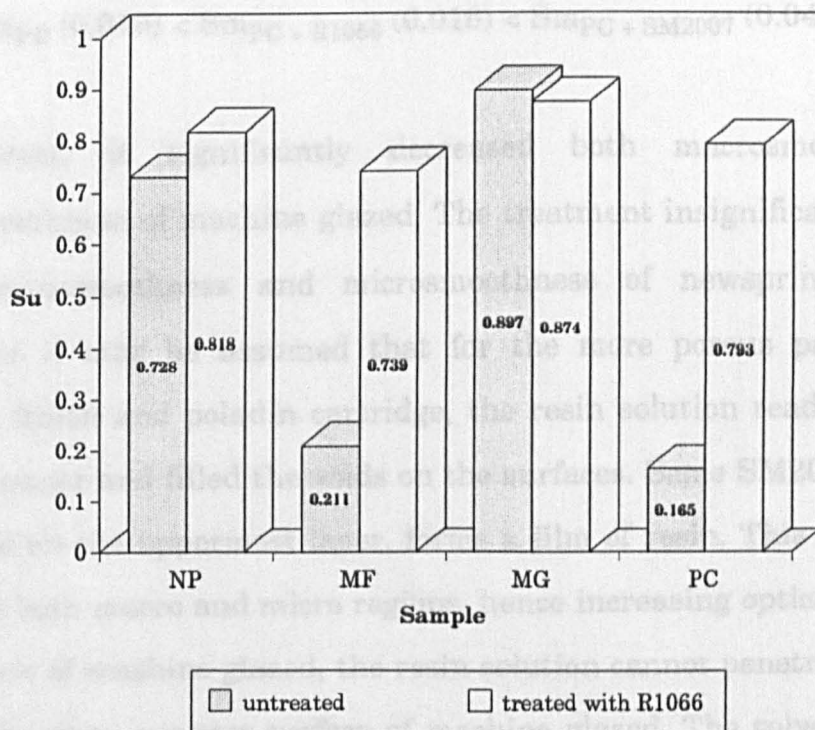
The optical smoothness results show the different behaviour of ink vehicles on paper. The effects of ink vehicles in changing the surface structure involve several aspects of fibre modification, such as fibre swelling, fibre relaxation or shrinkage of fibres. Changes in the smoothness parameters can be attributed to paper structure changes. This study also points to the role of surface structure in the penetration by each of the components.

The effects of individual ink vehicle components are most clearly illustrated by R1066 (Figure 3.6). It significantly increases both the macrosmoothness and microsmoothness of almost all the papers tested except machine glazed. These increases are more clearly seen in microsmoothness than those in macrosmoothness. In particular, the increases are greater for machine finish and poladin cartridge. In the case of machine finish, it is increased from a value of 0.211 to a value of 0.739 and in the case of poladin cartridge, it is increased from a value of 0.165 to a value of 0.793. Another obvious result is that the treatment results in a significant decrease in the macrosmoothness of machine glazed paper, the macrosmoothness is decreased from a value of 0.076 to a value of 0.042. This behaviour can be explained as follows. Owing to the high amount of furnish and sizing materials in machine finish, and to the coating mixture and the coating method used in poladin cartridge, it is envisaged that when R1066 penetrated into the paper, it may bring along these materials from the surface plane towards regions below the surface plane. This results in removing some microfeatures of the macro regions and, on the other hand, these materials deposit and may become compact on the micro regions of the

Figure 3.6 The effects of R1066 on (a) macrosmoothness and (b) microsmoothness of different types of paper.



(a)



(b)

surface. As a result, both the macrosmoothness and microsmoothness of machine finish and poladin cartridge will have increased significantly. On the other hand, R1066 may penetrate less into the more impervious surface such as that of machine glazed. This may cause the uppermost fibre layers of machine glazed to swell and debonding; and subsequently, after evaporation from the paper, the fibres shrink losing their compaction, resulting in a decrease in the macrosmoothness.

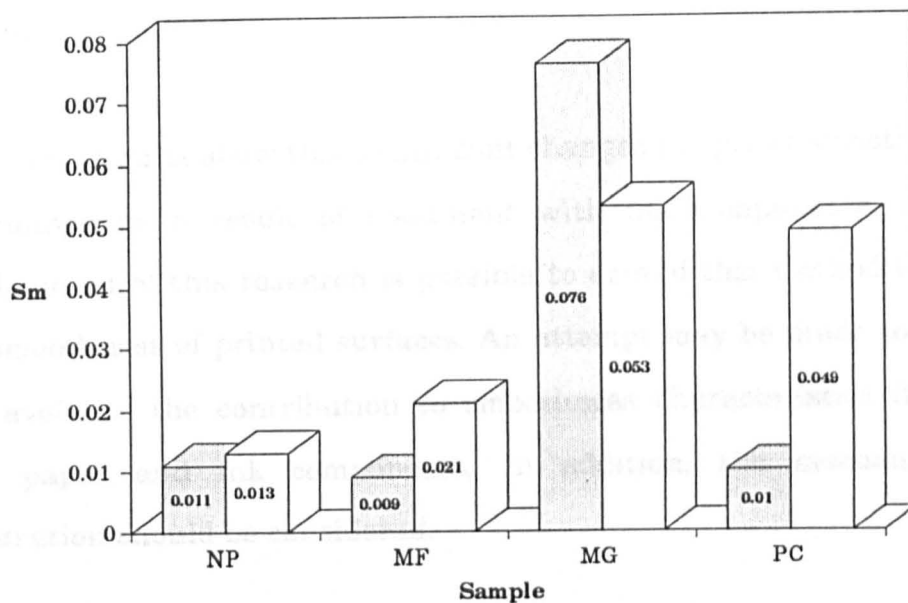
SM2007 represents a simple model of ink varnish. The effect of this vehicle (Figure 3.7) significantly increases both macrosmoothness and microsmoothness for machine finish and poladin cartridge. Moreover, it should be noted that the increases in the macrosmoothness of both machine finish and poladin cartridge treated with SM2007 are greater than those with R1066.

$$Sm_{MF} (0.009) < Sm_{MF+R1066} (0.015) < Sm_{MF + SM2007} (0.020)$$

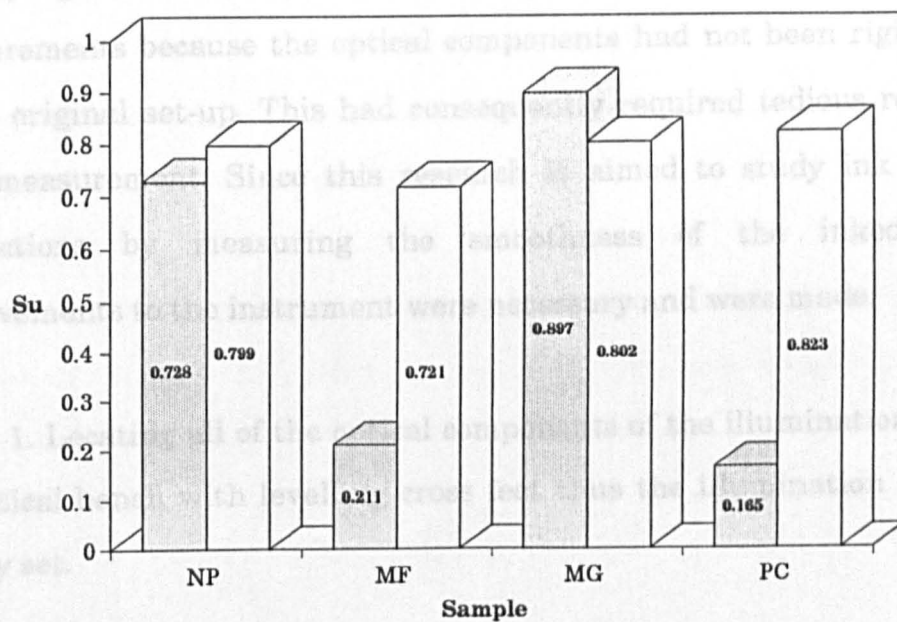
$$Sm_{PC} (0.010) < Sm_{PC + R1066} (0.018) < Sm_{PC + SM2007} (0.049)$$

In contrast, it significantly decreased both macrosmoothness and microsmoothness of machine glazed. The treatment insignificantly increases both macrosmoothness and microsmoothness of newsprint. From this behaviour it may be assumed that for the more porous papers such as machine finish and poladin cartridge, the resin solution readily penetrated into the paper and filled the voids on the surfaces. Some SM2007, which has remained on the uppermost layer, forms a film of resin. This creates mirror facets on both macro and micro regions, hence increasing optical smoothness. In the case of machine glazed, the resin solution cannot penetrate as a whole due to the more compact surface of machine glazed. The solvent component

Figure 3.7 The effects of SM2007 on (a) macrosmoothness and (b) microsmoothness of different types of paper.



(a)



(b)



may penetrate faster into the paper causing fibre swelling, relaxation and debonding which may in turn cause the film of the resin solution to follow the contour of the surface.

The results show that significant changes in optical smoothness can be determined as a result of treatment with ink components; and further development of this research is possible to extend this method to determine the smoothness of printed surfaces. An attempt may be made to distinguish and evaluate the contribution to smoothness characteristics derived from both paper and ink components. In addition, the mechanism of ink penetration should be considered.

### **3.6 IMPROVEMENTS OF THE INSTRUMENT.**

The preliminary study was aimed at quantitative measurements and in carrying out this work, there were some difficulties in making reflectance measurements because the optical components had not been rigidly located in the original set-up. This had consequently required tedious resetting for each measurement. Since this research is aimed to study ink and paper interactions by measuring the smoothness of the inked surfaces, improvements to the instrument were necessary and were made.

1. Locating all of the optical components of the illumination system on an optical bench with levelling cross feet thus the illumination system was rigidly set.



2. The former HeNe laser was replaced by a more reliable 5 mW linearly polarized HeNe laser Model 1125P from Uniphase Laser Ltd. and a power supply to operate this laser from Hughes Power Supply, Model 4020F.

3. A, modified, divided circle spectrometer was used to replace the previous mechanical stage. This classic instrument consists of three parts; a collimator and slit, a prism or grating on a rotating table and a viewing telescope on a rotating scale (Ealing Electro-Optics). Both the collimator and the telescope bases are heavy box form castings which make the instrument extremely stable and suitable for use as a mechanical stage. In order to achieve the improved functions of the mechanical stage, the main modifications of this instrument were as follows.

- the collimator arm was removed.
- the telescope was replaced with the photomultiplier.
- A graduated rotating table was modified to make the sample holder which is then fixed on top of the prism table. The prism table incorporated vertical control which enables the height of the sample holder to be adjusted to match that of the illumination system.

When all the improvements had been made and the new version of the instrument set up (Figure 3.8), the precision and repeatability of the instrument was evaluated.

1

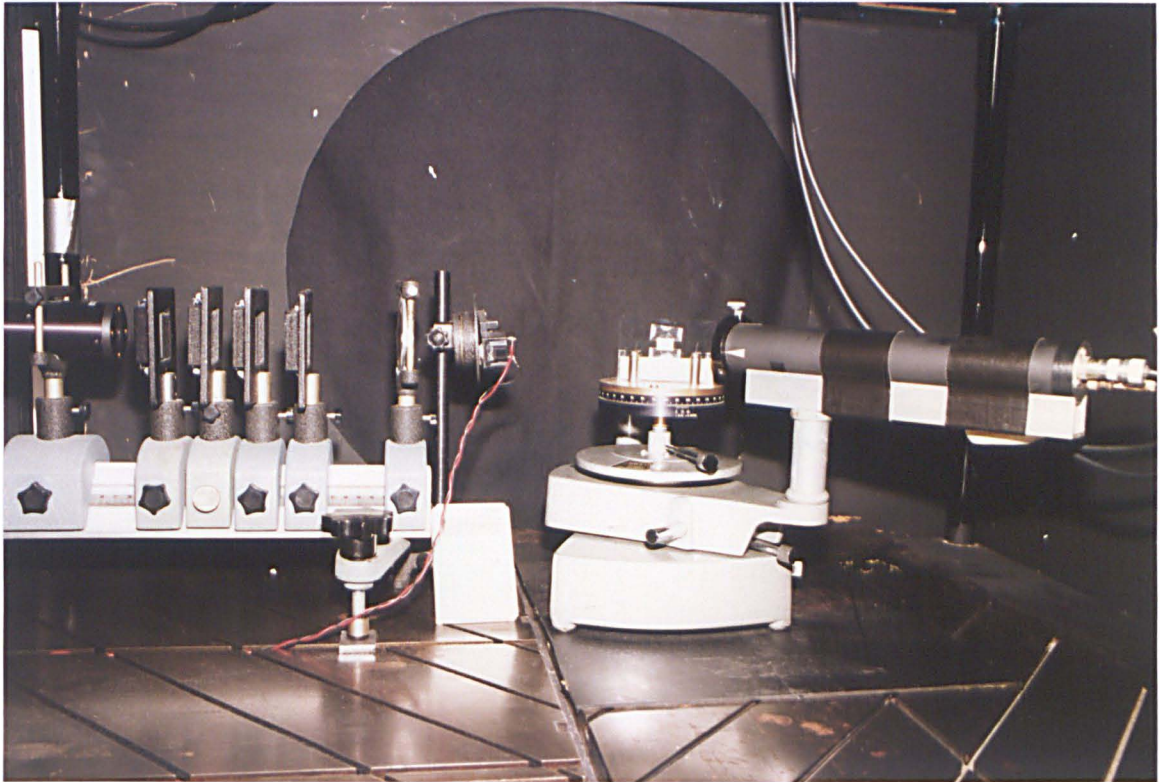
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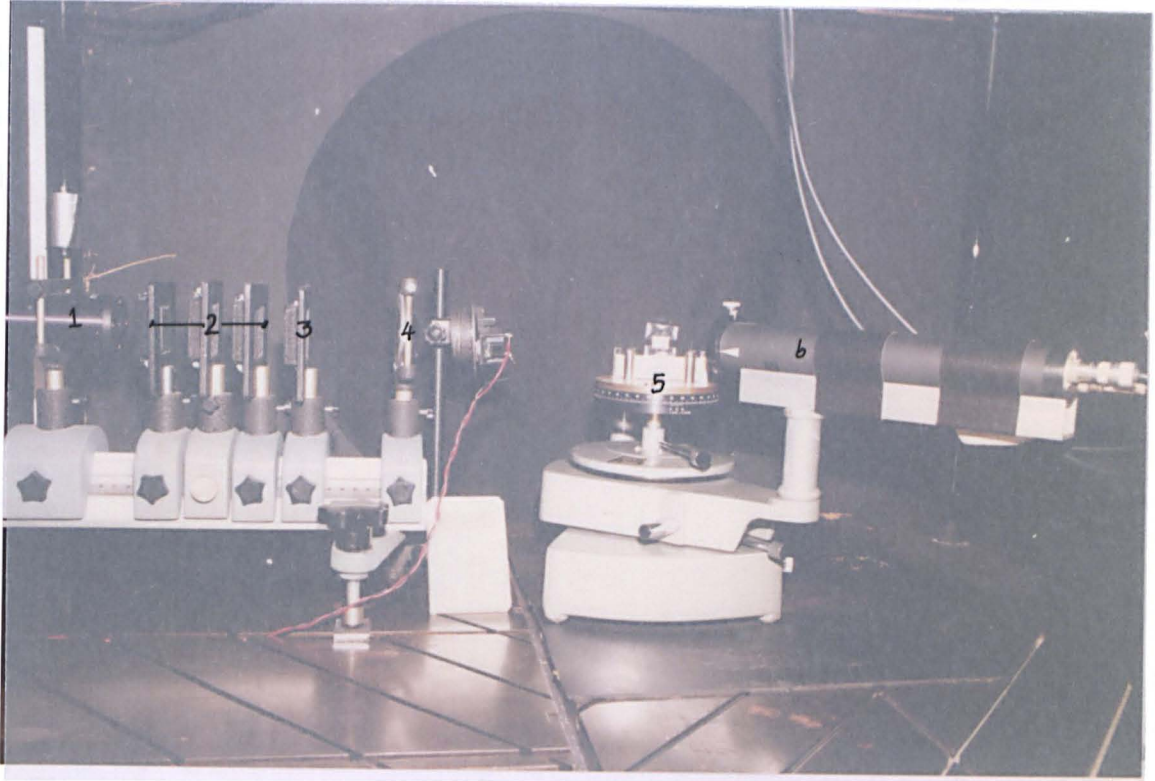
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6



**Figure 3.8 The optical reflectance instrument.**

- 1) A HeNe laser.**
- 2) Neutral density filters.**
- 3) A polarizer.**
- 4) A convex lens.**
- 5) A modified mechanical stage.**
- 6) A photomultiplier.**



**Figure 3.8 The optical reflectance instrument.**

- 1) A HeNe laser.
- 2) Neutral density filters.
- 3) A polarizer.
- 4) A convex lens.
- 5) A modified mechanical stage.
- 6) A photomultiplier.

### **3.7 REPEATABILITY OF THE INSTRUMENT.**

Comparing test results from different treatments may not indicate a quality difference if the difference between them can be attributed to errors arising from the instrument or from the operator. Thus, it is necessary to find the repeatability or the precision of the instrument employed in this study.

Repeatability refers to tests carried out under conditions that are as constant as possible in one laboratory by one operator and using the same instrument (BS 5497:Part1:1987, ISO 5725-1986). To obtain this, a set of ten reflectance measurements in a range of incident angles from 15 to 85 degrees was made on the standard glass. The data are presented in Table 3.5. The coefficient of variation gave a maximum value of 3.14 %.

Table 3.5 Repeatability of the reflectance instrument.

Measurement	Rs														
	15°	20°	25°	30°	35°	40°	45°	50°	55°	60°	65°	70°	75°	80°	85°
1st	4.3	4.7	5.3	6	7	8.2	9.8	11	14	18	25	32	42	56	74
2nd	4.4	4.9	5.5	6.2	7.2	8.4	10	11	14	18	25	32	42	56	74
3rd	4.4	4.7	5.4	6.1	7.1	8.2	9.8	11	14	18	24	32	42	56	74
4th	4.6	5	5.4	6.2	7.2	8.5	10	11	15	19	25	33	42	56	74
5th	4.6	5	5.5	6.2	7.2	8.5	10	11	15	19	25	33	43	56	74
6th	4.6	4.9	5.4	6	6.9	8.2	10	11	14	18	25	32	42	56	74
7th	4.6	5	5.4	6.2	7.1	8.1	9.8	11	14	18	24	32	42	56	74
8th	4.7	5.1	5.6	6.2	7.2	8.3	9.8	11	14	18	24	32	42	56	74
9th	4.7	5.1	5.5	6.3	7.2	8.2	10	11	14	18	24	32	42	56	74
10th	4.7	5.1	5.6	6.4	7.4	8.5	10	11	14	18	24	32	42	56	74
$\bar{X}$	4.56	4.95	5.46	6.18	7.15	8.31	9.92	11	14.2	18.2	24.5	32.2	42.1	56	74
S.D.	0.14	0.15	0.1	0.13	0.14	0.15	0.1	0	0.42	0.42	0.53	0.42	0.32	0	0
CV(%)	3.14	3.05	1.77	1.99	1.89	1.83	1.04	0	2.97	2.32	2.15	1.31	0.01	0	0

#### **4. STUDY OF INKED SURFACES.**

##### **Evaluation of printed image characteristics by an optical method with particular reference to offset lithographic printing.**

The quality of a solid print has been characterized so far by density measurements [Williams, 1988; Visa & Langinmaa, 1992]. Little work has been described for detailed laboratory methods capable of evaluating prints in terms of print smoothness. The present study is directed towards the use of the non-contact optical method [Hansuebsai, 1989] to gain further insights into the characteristics of printed surfaces which are the results of material interactions in the printing process. The technique has been reviewed and the optical reflectance instrument improved to allow the measurement of solid printed surfaces. The reflectance data were used to calculate both macrosmoothness and microsmoothness for the printed surfaces. Other related methods have been reported. Gate & Parsons [1993]<sup>\*</sup> have reported further improvements in the optical reflectance examination of coated papers; Mangin [1993] has demonstrated the use of confocal laser microscopy in determining 3D profiles of a paper surface, moreover, he obtained, simultaneously, information on the microscopic distribution of the ink film by incorporating a fluorescent dye in the ink. Extensive information about ink, paper and water interactions has been published [e.g., Festko, 1986; Trollås, 1987; Aspler & Lepoutre, 1991], however, little work has been done on how the presence of water or fountain solution emulsified in the ink may influence both ink transfer and setting which in turn determine the printed surface characteristics. Thus the effects of emulsification, i.e., fountain solution emulsified in the ink, on print smoothness are investigated.

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<sup>\*</sup> This paper is important as much of this work is paralleled to this study and complements the work presented here.

## 4.1 MATERIALS.

**4.1.1 Paper samples.** Two different types of printing papers; an uncoated and a coated type, were chosen in this study. They were expected to interact differently with the inks due to their variation in properties. Hence, the measurements were expected to reflect within limits ink and paper interactions and the effects of fountain solution on printed surface characteristics. Suprawhite machine glazed poster was chosen to represent the uncoated papers and Hispeed gloss art to represent coated papers. Machine glazed paper and gloss art paper will be referred to in Tables and Figures as MG and GA respectively.

*Machine glazed paper (MG).* This was the same type as used in the preliminary study.

*Gloss art paper (GA).* High quality printing normally requires a very smooth surface. Since supercalendering of uncoated papers alone gives a limited smoothness, coating is an excellent means of improving the surface of paper in term of both printability and optical properties. When compared with uncoated papers, one of its greatest contributions to print quality is its excellent affinity for printing inks [Franklin, 1970].

Gloss art is a coated paper having a high gloss surface which is achieved by the application of more than one layer of coating on the base paper. Paper coatings are mixtures of pigments in a liquid solution of an adhesive or binder to hold the pigment to the base stock. These layers can be applied at different stages of the paper making process to fill in the surface voids of the base paper thus producing a more uniform smooth surface which



is more uniformly wetted by printing inks. If a highly smooth surface is required then this is achieved by supercalendering. The amount of supercalendering to obtain a suitable printing smoothness depends on the evenness and the amount of coating.

The paper samples were stored in the laboratory room which was used for optical smoothness measurements and were cut into approximately 4.5 cm by 30 cm. strips to be used with the IGT printability tester. It should be noted that throughout the course of this study, the same batch, same side and same machine orientation was used for each paper. The properties of the papers are given in Table 4.1.

Table 4.1 The paper properties of gloss art paper and machine glazed.

Property	GA	MG
Grammage ( $\text{g} / \text{m}^2$ )	114.9	90.4
Calliper (mm)	0.45	0.65
Bendsten porosity (ml / min)	5	875
Parker Print-surf roughness (microns)	0.75	4.73
K&N oil absorption test ('K&N' unit)	14.6	54.9
Cobb water absorption test ( $\text{g} / \text{m}^2$ )	181	131
Gloss (%)	35.5	8.3

Paper tests were carried out by the following methods in a laboratory maintained at  $23^\circ\text{C} \pm 1^\circ\text{C}$  and  $50\% \pm 2\%$  RH.

The grammage of paper was determined on a quadrant balance and an analytic balance.

The calliper of paper was obtained using a dead-weight dial micrometer.

The surface roughness and the porosity were evaluated by air-leak methods. The surface roughness was measured using a Parker-Print-surf with soft backing and pressure loading at 10 kgf. The porosity was measured on the Bendtsen-type instrument, with standard weight 150 grams.

The ink absorbency characteristics were determined by use of K & N ink.

The water absorbency characteristics were obtained using the Cobb size test.

The specular gloss was measured on a Minigloss meter from Sheen Instruments which complies with DIN 67530, ISO 2813, BS 3900-6161. According to DIN 67530, a measurement angle of 20° is used for high gloss surfaces, 60° for medium-gloss surfaces and 85° for dull surfaces. A measurement angle of 60° was chosen in this study.

**4.1.2 Ink sample.** Since the optical reflectance instrument employs a HeNe laser 632.8 nm as the light source, the choice of inks to be studied was restricted to yellow or magenta inks whose reflectance spectra are high at the emission wavelength of HeNe laser. A commercial heatset yellow ink was thus chosen for this study and was supplied by Coates Lorilleux International. This ink sample will be referred to as Y-ink.

**4.1.3. Fountain solution.** The main function of the fountain solution is to keep the non-image area clean and free from ink in order to obtain a clean

print. For this reason, the main active component of any fountain solution is the desensitiser composed of strong hydrophilic colloids such as gum arabic, water, and activated with acid such as phosphoric acid.

The addition of alcohol, usually isopropanol, is made to the fountain solution. Quantities used are generally between 10% and 15% [Rosos, 1990]. Braun [1985] reported that isopropanol promotes the formation of emulsion with finer and more uniform droplets of fountain solution in the ink. Moreover, alcohol evaporates more readily from the plate surface leading to the following: less emulsification, quicker establishment of ink and water balance, better drying of the inks and better trapping of ink onto paper.

A pure grade (99.7%) of isopropanol (or 2-propanol) from John Seaton Ltd. and deionized water from the Seradest S600 - Seral Water Purification System, in the proportion of 1:9 by weight was formulated to be the fountain solution model. It was claimed that 10% isopropanol is the lower limit of effective isopropanol concentration in the fountain solution [Fadner, White & Hayden, 1976]. This fountain solution will be referred to as F.S..

## **4.2 EXPERIMENTAL METHOD.**

**4.2.1 Pre-emulsified ink preparation.** Bassamir & Krishnan [1991] stated that a widely used procedure to simulate offset lithographic printing is the use of pre-emulsified inks. A fixed amount of fountain solution is emulsified into the ink using a high speed stirrer on a laboratory mixer before printing. The crucial stage is that the 'emulsion ink' must be transferred very quickly to avoid loss of the emulsified fountain solution.

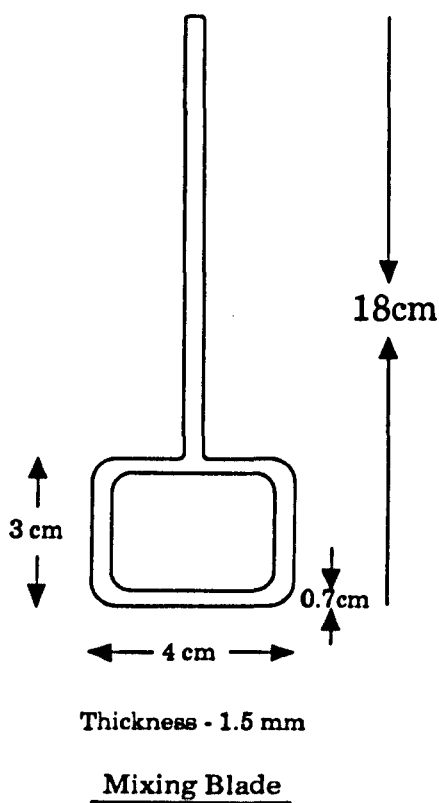
*Apparatus.* The apparatus described below was used for both the pre-emulsified ink preparation and the emulsification rate tests which will be discussed later.

1. A high speed laboratory stirrer with speed control and a drill chuck to take the paddle, supplied by Janke & Kunkel GMBH & Co.

2. A single paddle mixing blade (Figure 4.1). This is a modified mixing blade used in Coates Lorilleux International.

3. A beaker of approximately 90 mm external diameter. The base should be flat and the sides perpendicular to allow efficient mixing.

4. An analytic balance.



**Figure 4.1 A modified mixing blade.**

### *Procedure.*

1. A clean empty beaker was placed on the balance and the balance was tared.

2. 90 g of yellow ink was weighed into the beaker.

3. 10 g of fountain solution was added to the ink to make 100 g of 'emulsion ink'. This can be described in terms of 10% water content 'emulsion ink'.

4. The paddle was connected to the stirrer.

5. The stirrer speed was adjusted to 225 rpm at the shaft. The ink and fountain solution was mixed at this speed for 5 minutes. Then the stirrer speed was increased to 350 rpm and the emulsion was mixed for a further 3 minutes. The beaker should be rotated about its base during stirring to ensure complete mixing.

6. After mixing, the 'emulsion ink' was put in a clean air-tight can. It was assumed that the evaporation of fountain solution was very little and could be considered negligible.

7. Steps 2 to 6 were repeated for another two different proportions of ink and fountain solution; 80g of ink with 20g of fountain solution making approximately 20% water content and 70g of ink with 30g of fountain solution making approximately 30% water content.

It should be noted that the term water is used generically to mean the fountain solution. The relationship between water pickup (WP) and water content (WC) can be written as  $WP = WC / (100-WC)$ . Table 4.2 shows the conversion of water content values into water pickup values.

Table 4.2 The relationship between water content and water pickup.

Water content (%)	Water pickup(%)
10	11.1
20	25.0
30	42.9

**4.2.2 Printing.** The ink and the corresponding 'emulsion inks' were printed using an IGT printability tester, model AIC 2-5. The IGT printability tester is a precision, press-like testing device, i.e., printing speed, pressure and inking can be adjusted with the required precision. These conditions can be varied independently so that the effects of printing conditions on prints can be evaluated. The IGT printability tester consists of two separate units: an inking unit with an ink pipette for accurately measuring small quantities of ink; and a printing unit. Each 'ink' was distributed in the inking unit for two minutes, before inking the printing disc for thirty seconds. The inking unit and the printing disc were cleaned between each transfer measurement. It is claimed by the IGT manufacturer that the design of the inking unit ensure that the distribution of ink over the inking assembly does not cause an increase in temperature. Thus, it is believed that the inking procedure, used as a standard inking procedure in this study, would lead to a standard minimized loss of emulsified fountain solution before the ink is transferred to the paper. The printing throughout this study has been carried out under controlled printing conditions, with a rubber covered printing disc of 32 mm width, to simulate offset lithographic printing. The pressure applied was set at 60 kgf and the printing speed at 1 m/s.

In practice, a printer is interested in printing to a standard print density, therefore the comparison is made at a constant print density rather than at a constant amount of ink on paper. Also prints are generally made to obtain a solid print density to suit the type of paper. The British Printing Industries Federation (BPIF) provides typical averages of solid density as given in Table 4.3 for greater standardisation when printing colour work by the offset lithographic process.

Table 4.3 The BPIF recommendation for solid densities.

<b>Paper</b>	<b>Cyan</b>	<b>Magenta</b>	<b>Yellow</b>	<b>Black</b>
Art paper	1.4	1.4	1.0	1.8
Matt coated paper	1.3	1.3	0.95	1.7
Uncoated paper	1.15	1.1	0.9 <sup>a</sup>	1.5

Tolerances are of the order of  $\pm 0.1$ .

a - Gretag standard value = 0.85.

In the present study, densities were measured using a Gretag densitometer, model D-186. Gretag has also proposed standard values for the solid densities. The value for yellow density is very close to those of the BPIF (See footnote of Table 4.3). The density levels of the test prints were set at  $D \sim 0.92$  for gloss art and  $D \sim 0.84$  for machine glazed. The variations between the 'inks' were largely attributable to the variations in the water content of the 'inks', thus it was found necessary to transfer differing amounts of 'inks' to each paper sample to achieve the target density. This enabled the influences of fountain solution to be examined. The amounts of

ink transferred to the paper as shown in Table 4.4 were determined by weighing the printing disc before and after printing. The weights were divided by the surface area of the inked image and reported as weight per unit area ( $\text{g/m}^2$ ).

Table 4.4 The amounts of ink transfer for gloss art prints and machine glazed prints.

Sample	ink transferred ( $\text{g/m}^2$ )
Gloss art (D~0.92)	
GA + 100% Y- ink	0.77
GA + 90% Y- ink : 10% F.S.	0.81
GA + 80% Y- ink : 20% F.S.	0.84
GA + 70% Y- ink : 30% F.S.	0.89
Machine glazed (D~0.84)	
MG + 100% Y- ink	1.55
MG + 90% Y- ink : 10% F.S.	1.57
MG + 80% Y- ink : 20% F.S.	1.59
MG + 70% Y- ink : 30% F.S.	1.61

**4.2.3 Drying.** Since the 'inks' used were heatset inks, each printed strip was dried immediately after printing by passing through a Sinvatrol oven, a heatset tester oven . The heat was provided by 2 hot air blowers which enable the temperature of the oven to be set. The belt speed was set at 6 m/min and the oven temperature was set at 150 °C.



**4.2.4 Rheological property measurements.** The rheological properties of the ink and the corresponding 'emulsion inks' are given in Table 4.5.

*Viscosity measurements.* They were made on a Haake VT viscometer, a 'cone and plate' viscometer. The viscosity was determined by measuring the torque required to rotate a cone over a flat plate, with the liquid sample placed in between. The viscosity is normally determined at two speeds; speed 1 (high shear) and speed 4 (low shear). However, when reporting viscosity measurements of a sample, the reading at speed 1 is quoted.

*Tack measurement.* It is a measure of the force required to split the ink film and tack measurements were carried out on a Tack-o-Scope.

Table 4.5 The rheological properties of heatset yellow ink and the corresponding 'emulsion inks'.

Property	Ink sample			
	100%Y-ink	90%Y-ink +10% F.S.	80%Y-ink +20% F.S.	70%Y-ink +30% F.S.
Viscosity, (Poise) 32°C	80.6	41.5	34.6	29.3
Tack, ('Tack-o-Scope' unit), 25°C	113	110	105	108

**4.2.5 Evaluation of printed surface characteristics.** For solid printing, the most important print qualities are print smoothness and print gloss.

*Print smoothness.* Unprinted paper samples (both machine glazed paper and gloss art paper) and the printed samples were evaluated by the optical method described in the preliminary study. Twenty points were evaluated from each sample and the results are given in Appendix E (Tables E.1-E.4).

*Print gloss.* Print gloss was measured after the printed strips were dried for at least 24 hours. Gloss readings of both unprinted and printed samples were taken on a Sheen Minigloss, model 101 at an angle of 60°. This instrument was designed to be used only on flat surfaces and used to measure specular gloss. Readings were averaged for the final gloss values and the results are presented in Table 4.6.

Table 4.6 Gloss values of unprinted and printed samples for both gloss art and machine glazed.

Sample	Gloss (%)
Unprinted GA	35.5
GA + 100% Y- ink	57.6
GA + 90% Y- ink : 10% F.S.	59.1
GA + 80% Y- ink : 20% F.S.	59.5
GA + 70% Y- ink : 30% F.S.	53.3
Unprinted MG	8.3
MG + 100% Y- ink	8.1
MG + 90% Y- ink : 10% F.S.	8.2
MG + 80% Y- ink : 20% F.S.	8.1
MG + 70% Y- ink : 30% F.S.	8.0

**4.2.6 Data analysis.** The optical smoothness results were subjected to statistical methods such as t-test, analysis of variance and Multiple comparison test.

The t-test was used to test whether the optical smoothness mean values of two sample groups are the same. The test is based on the means and the variances of both the samples and the theoretical population under consideration. The calculated t-value was compared to the values of the t-distribution with  $(n_1 + n_2 - 2)$  degrees of freedom and at 0.05 significance level where  $n_1, n_2$  are the numbers of cases in each sample group. These are the critical values of T. A significant difference between these two samples was obtained when

a) the calculated t-value was equal to or greater than the (positive) critical value of T.

or b) the calculated t-value was equal to or less than the (negative) critical value of T.

Analysis of variance and Duncan's multiple comparison test were employed to test the optical smoothness results of several sample groups as previously described in the preliminary study.

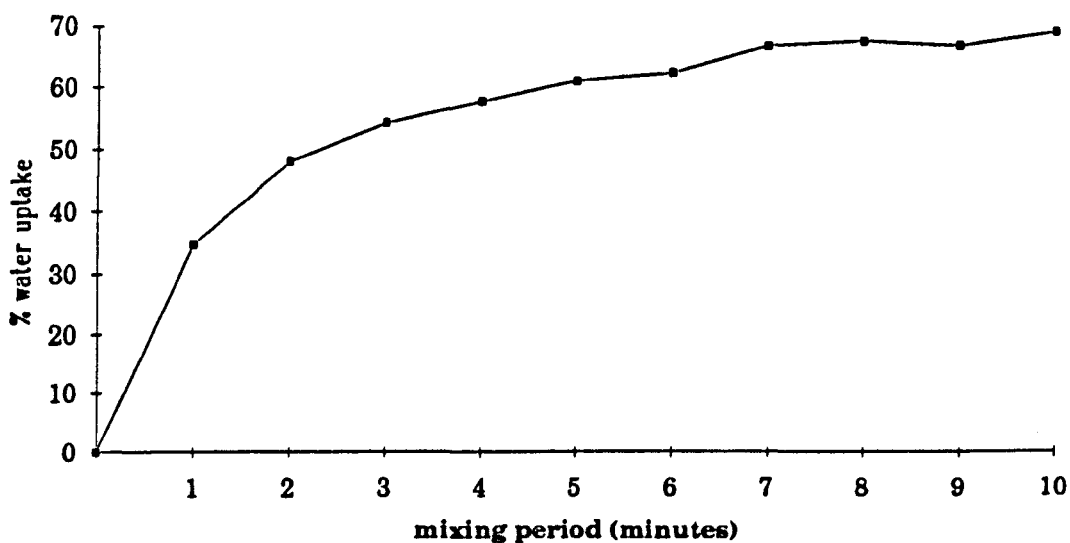
The results from these statistical methods are reported in Appendix F (Tables F.1-F.5).

**4.2.7 Water pickup or emulsification rate test.** Measurement was carried out with a modified Surland test using the same apparatus as in the pre-emulsified inks preparation stage. The procedures are explained below.

1. A clean empty beaker and the mixing paddle were placed on the balance and the balance was tared.

2. 50 g of ink was weighed into the beaker and the total weight of the beaker, stirrer paddle and ink recorded.

3. 50 g of fountain solution was added to the ink.
4. The stirrer speed was adjusted to 400 rpm at the shaft.
5. The paddle was connected to the stirrer and the ink and fountain solution mixed for a period of one minute. The beaker should be rotated about its base during mixing to ensure complete mixing.
6. After one minute, the beaker, stirrer and contents were removed and any excess fountain solution was poured off. Any trapped fountain solution that was not in a refined emulsion was separated from the bulk by very gently mixing with the paddle for not more than one minute and pouring away that fountain solution. The beaker, stirrer and contents were then reweighed and the difference in weight of fountain solution recorded.
7. Step 5 and 6 were repeated for a further nine x one minute periods. 50 g excess fountain solution should be maintained throughout the test.
8. A graph was plotted of water pickup versus time, Figure 4.2.



**Figure 4.2 The percentage water uptake of heatset yellow ink.**

**4.2.8 Scanning electron microscopy (SEM).** This is a very useful instrument to give an image of a surface. An electron beam is used to illuminate the sample. When the sample is exposed to the scanning electron beam, low energy secondary electrons are released from the sample surface, collected and transmitted to a cathode ray tube to form an image, The image is then photographically recorded.

As the angle of the electron beam is stationary, the angle in which the beam strikes the sample surface is dependent on the tilt angle of the sample. It is known that tilt angles of the sample between 30° to 45° are the best conditions for observing porosity and micro detail [Settlemyer, 1992]. In this study, unprinted and printed samples were observed by scanning electron microscopy. This was carried out on a Hitachi scanning electron microscope, model S-450. The tilt angle of the sample was set at 45°. This SEM method is not influenced by the colour of the printing ink. However, at high resolution the ink pigment can be observed since their particle shape is significantly different from that of the fibres. The aim of employing SEM was to provide a better understanding of the surface characteristics and provide independent evidence for the surface characteristics.

#### 4.3 RESULTS AND DISCUSSION.

This work has developed through four stages. Firstly, the optical method used has been developed by Hansuebsai [1988, 1989]; secondly, that method was improved and developed so as to be able to assess printed samples; thirdly, ink and paper interactions have been studied with general reference to the printing process; and fourthly, the effect of fountain solution in offset lithographic printing has been explored. The results obtained are conveniently presented and discussed in two parts.

In the brief results sections the amount of data involved is such that only the overall summaries will be presented in this chapter; the detailed summaries of the results are presented in the Appendices bound with this thesis; and the raw data itself comprises an extensive set of tables from which the Appendices have been derived.

Moreover, in view of the mass of data required, it was decided to restrict the paper examples to two contrasting types namely, gloss art and machine glazed.

*Part I. Evaluating unprinted and printed paper surface characteristics:* A criterion for quality of print is proposed in terms of print smoothness. Printed surface characteristics are discussed in relation to ink and paper interactions. This part will also highlight the contributions of paper properties to the characteristics of printed surfaces.

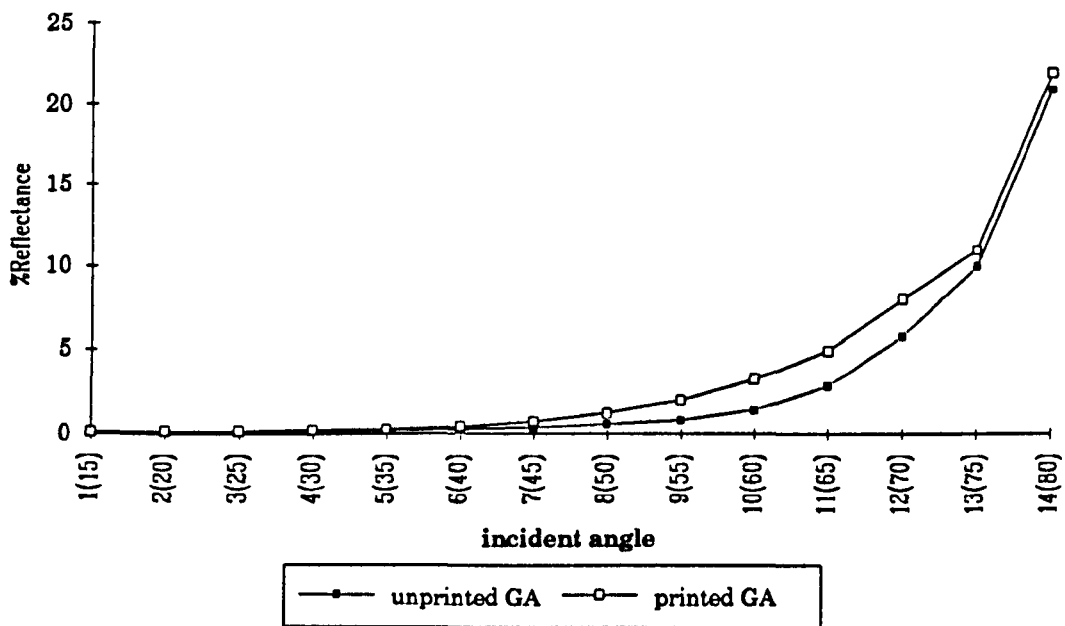
***Part II. Studying the effects on print smoothness:*** Fountain solution and ink which are the major additional factors affecting print smoothness in the offset lithographic process.

**Part I. Printed paper surface formation - Print smoothness consideration.**

**4.3.1 Results - Part I.**

From the preliminary study, the reflectance instrument has been improved and thereafter employed to evaluate printed surfaces. In the present (Part I) study, prints were prepared using two different types of paper printed with a heatset yellow ink on a laboratory IGT printability tester. It was also found from the preliminary study that the number of samples examined was not sufficient to draw precise conclusions. Paper testing practice is generally carried out with a minimum of ten samples (BS 3430:1986, ISO 186-1985). However, since the study aims to investigate the effects of factors in the offset lithographic process, the number of samples had to be increased to twenty samples (which is appropriate within the time frame of this study); this was found to be necessary so as to increase the sensitivity of the method and the reliability of the results. Moreover, the application of statistics to the results can enable conclusions to be drawn within more precise limits in terms of a definite probability (eg. 95% Confidence Limits). As a result, during the course of this study, more than two hundred sets of measurements were undertaken which also produced more than two hundred optical smoothness results. All results cannot be presented in this thesis thus it was decided to choose one sample from each group to represent its property. These examples are given in Appendix G.

This optical method involves surface reflection measurements and statistical correlation and regression analysis of the reflectance data to yield smoothness parameters. Figure 4.3 shows the reflectance characteristics from one random sample each, of unprinted and printed gloss art. It can be seen that the reflectance characteristic curve of the printed samples has a similar shape to that of the unprinted sample.



**Figure 4.3 An example of the reflectance characteristic of unprinted and printed gloss art.**

Table 4.7 summarizes macrosmoothness and microsmoothness, for each pair of unprinted and (corresponding) printed samples. A greater value of smoothness corresponds to a smoother surface. The macrosmoothness mean value of printed gloss art is greater than that of unprinted gloss art; the macrosmoothness mean value of printed machine glazed is, however, lower than that of unprinted machine glazed. Nevertheless, the results from



t-test statistical analysis revealed that the only significant difference was that between the macrosmoothness of machine glazed printed with heatset yellow ink and that of unprinted machine glazed. Therefore, the printed surface characteristics of gloss art print can be considered to be as smooth as the paper surface characteristics of unprinted gloss art since no significant differences were observed for both macrosmoothness and microsmoothness. - For the case of machine glazed, despite the absence of significant differences in the microsmoothness values, because of the lower value of its macrosmoothness, its printed surface characteristics can be considered to be rougher than the paper surface characteristics of unprinted machine glazed. These optical smoothness comparisons are illustrated in Figures 4.4-4.5.

The scanning electron micrographs of printed samples for both gloss art and machine glazed as compared with unprinted samples are given in Figures 4.6-4.7 respectively. The micrographs of both prints clearly illustrate a distinct layer of ink covering the paper surface.

Table 4.7 Optical smoothness summary: unprinted and printed gloss art and machine glazed samples. Each result given is the average value of twenty different points for each sample (See Appendix E ).

(a) Macrosmoothness (Sm)

Sample	Sm ( $\bar{X}$ )	S.D.	95% C.I. for $\bar{X}$	Min.	Max.
Unprinted GA	0.968	0.074	0.934 to 1.003	0.873	1.129
GA + 100% Y- ink	0.974	0.084	0.935 to 1.014	0.855	1.133
Unprinted MG	0.111	0.020	0.101 to 0.120	0.079	0.154
MG + 100% Y- ink <sup>a</sup>	0.089	0.015	0.082 to 0.096	0.071	0.126

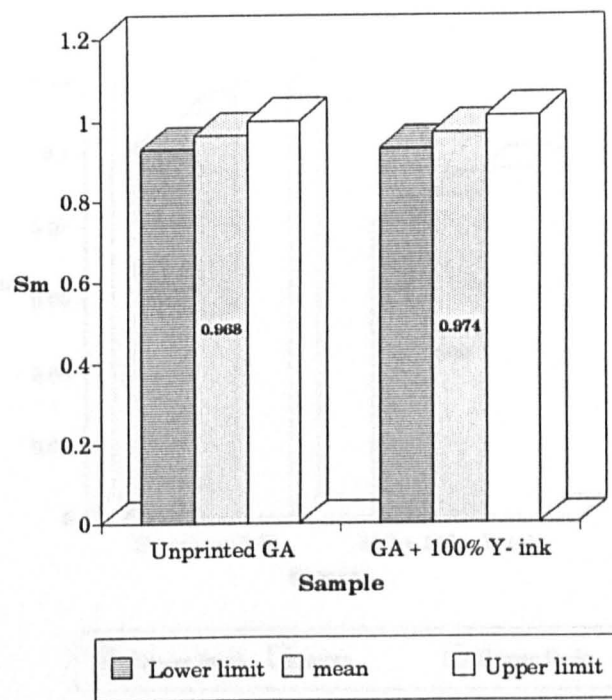
a - Sm was significantly different when compared with unprinted MG (See Appendix F and Figure 4.5).

(b) Microsmoothness (Su).

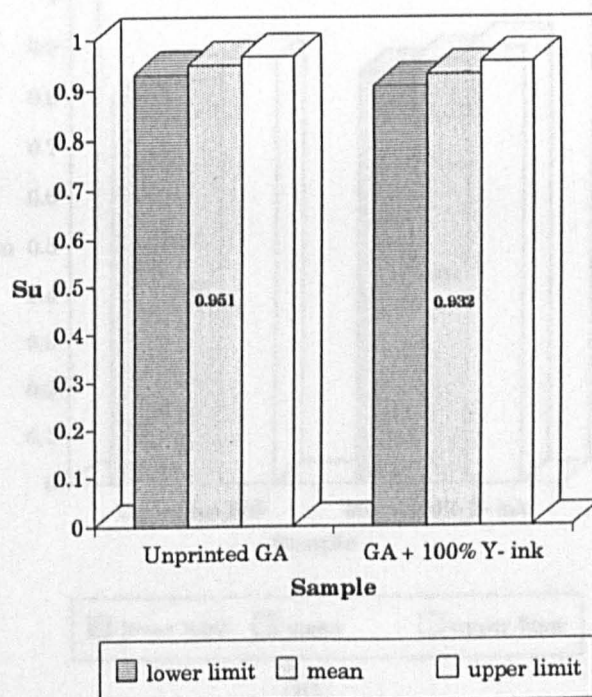
Sample	Su ( $\bar{X}$ )	S.D.	95% C.I. for $\bar{X}$	Min.	Max.
Unprinted GA	0.951	0.037	0.933 to 0.968	0.872	0.991
GA + 100% Y- ink	0.932	0.052	0.908 to 0.957	0.785	0.995
Unprinted MG	0.837	0.079	0.800 to 0.874	0.677	0.951
MG + 100% Y- ink	0.874	0.056	0.848 to 0.901	0.750	0.958

No significant differences were observed.

Figure 4.4 Optical smoothness of gloss art.  
 (a) Macrosmoothness.  
 (b) Microsmoothness.



(a)

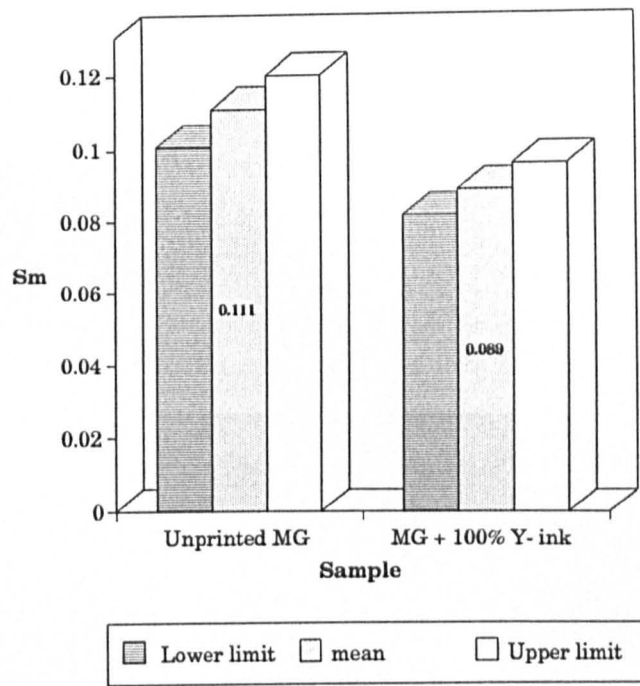


(b)

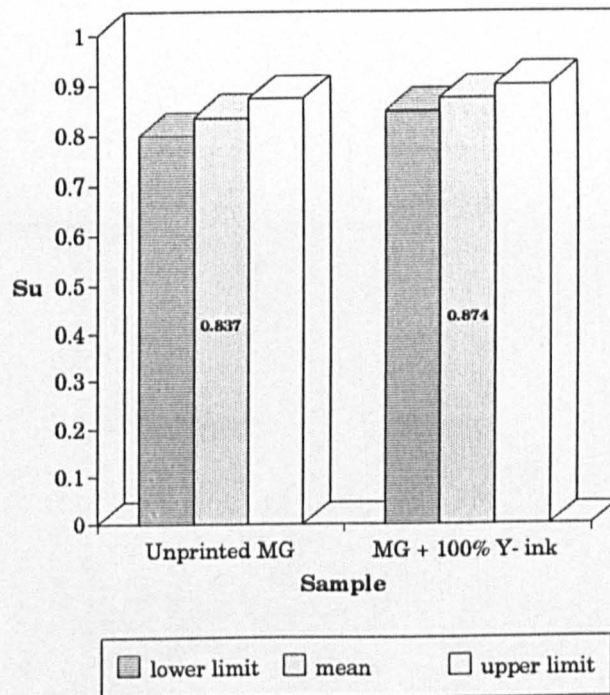
Figure 4.5 Optical smoothness of machine glazed.

(a) Macrosmoothness.

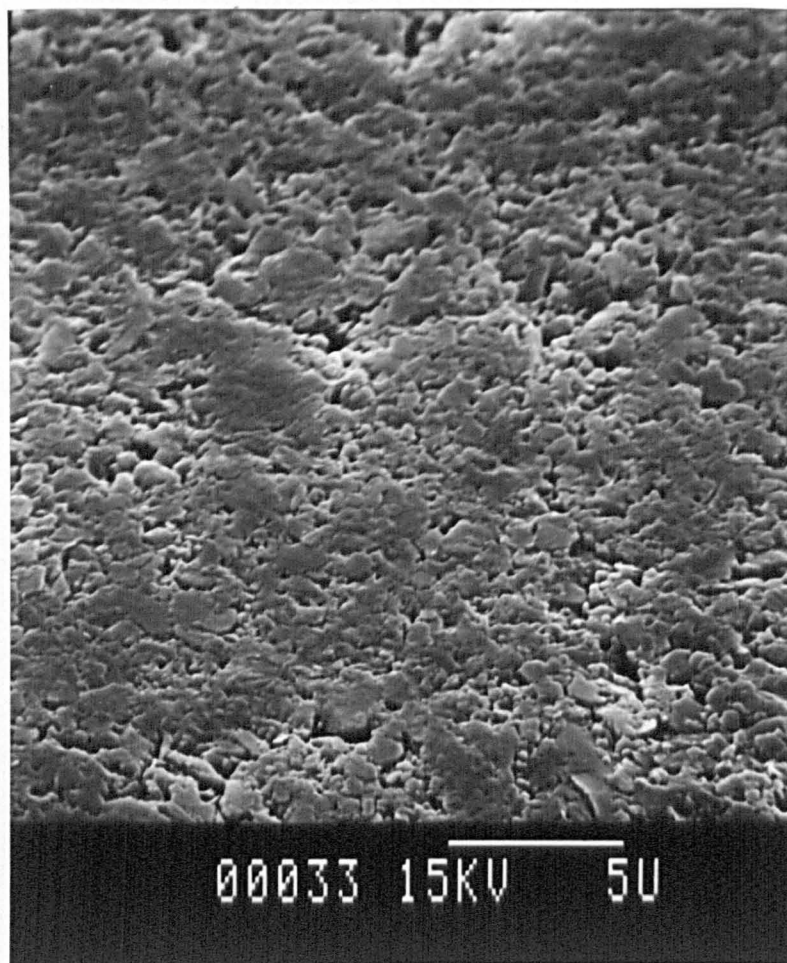
(b) Microsmoothness.



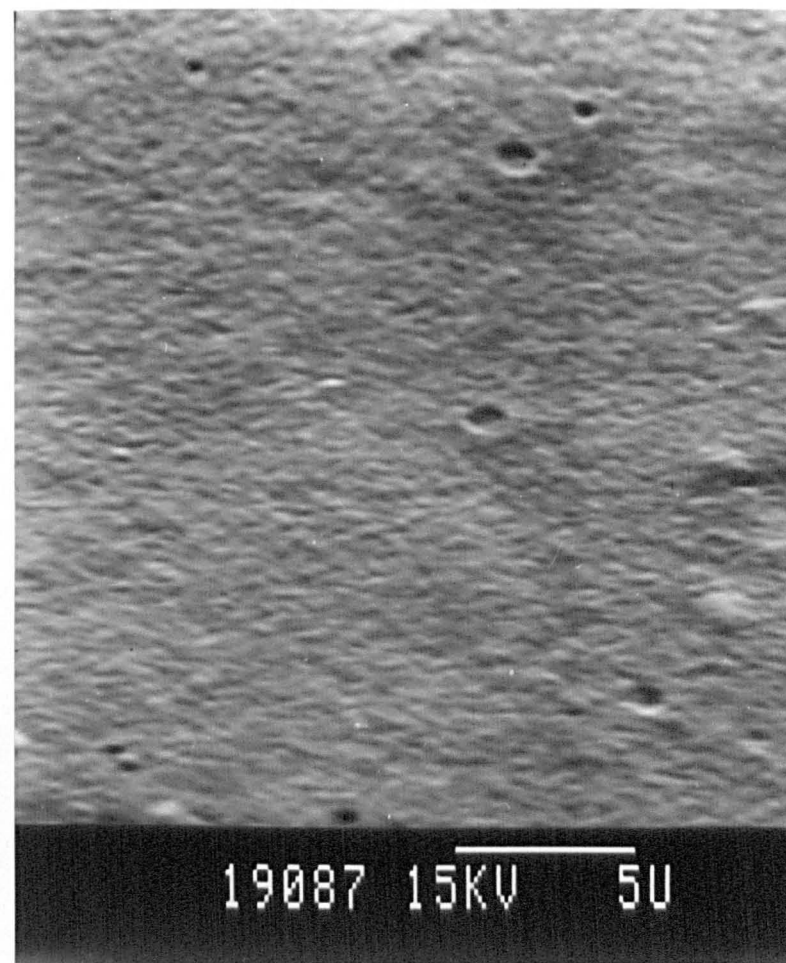
(a)



(b)



(a)



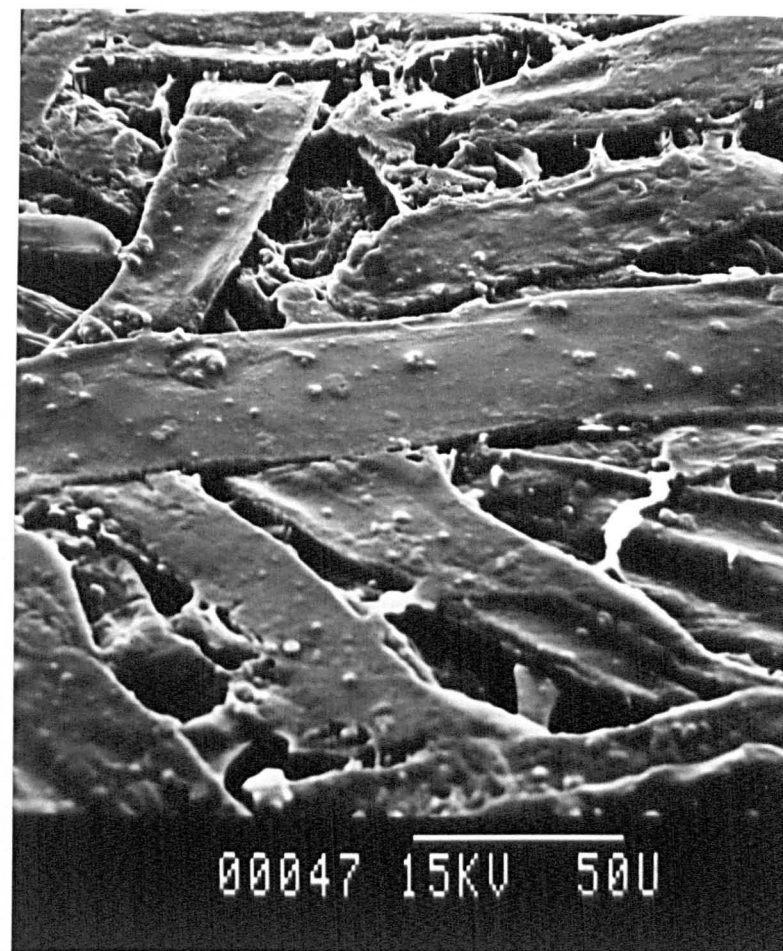
(b)

**Figure 4.6 Scanning electron micrographs x 5000.**  
**(a) unprinted GA.**  
**(b) GA printed with 100%Y-ink.**





(a)

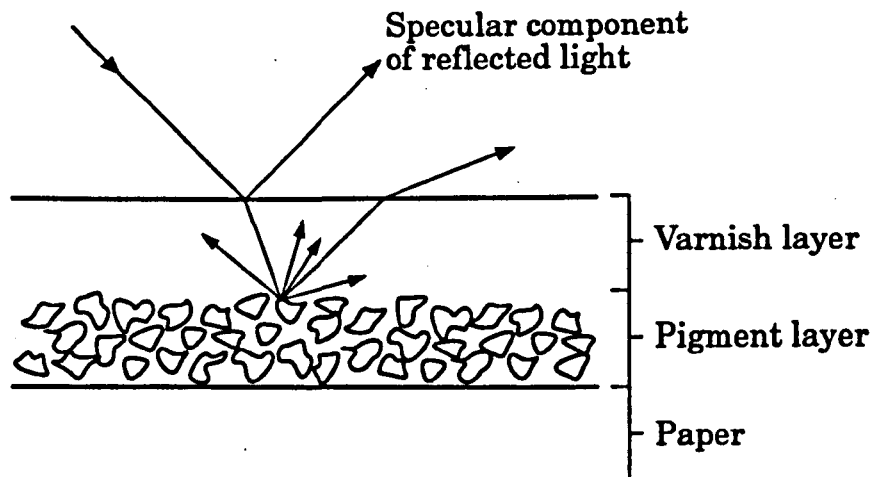


(b)

**Figure 4.7 Scanning electron micrographs x 600.**  
**(a) unprinted MG.**  
**(b) MG printed with 100%Y-ink.**

### 4.3.2 Discussion - Part I.

An ink is generally composed of pigments, ink vehicle and additives. The proportion of pigments in most inks is approximately 10-15% (that of the yellow pigment in the heatset yellow ink is 12.8%), therefore the main constituent is the ink vehicle. Moreover, it is generally known that one important factor contributing to the smoothness of the ink film is the amount of the ink vehicle 'holdout' on the paper surface. This means that if the pigment particles are completely covered by a level film of the vehicle, the ink film appears smooth. Therefore, a printed surface model is proposed as consisting of an ink vehicle-air interface rather than a pigment-air interface. This ink vehicle-air interface makes the ink film not far removed from Barkas' classical model, according to which any small area of the ink film is equivalent to a construct of mirror facets and rough facets. Mirror facets which are related to macro elements such as the dried polymer film of ink vehicle will reflect the incident light specularly. On the other hand, rough facets which are related to micro elements such as the pigment particles within the dried film will scatter the light. In addition, highly inclined mirror facets will lead to multiple reflections between sets of such facets. This is equivalent to diffuse reflection. Consequently, the reflection from the ink film includes surface specular reflection, surface scattering reflection and re-emerging diffuse reflection as shown in Figure 4.8.



**Figure 4.8 Reflection from the ink film.**

The method uses Fresnel's reflectance data, averaged for a range of refractive index values of 1.5 - 1.9 as the reference standard data (See Appendix B); the same reference standard applies for both unprinted and printed surfaces owing to the similarity in the magnitude of the refractive index values of paper and ink. The refractive index of paper is 1.55 [Barkas, 1939]; that of the oil-based vehicle is approximately 1.45-1.65 [Judd & Wyszecki, 1976; Young, 1973].

Hemstock [1962] stated that Mie theory is not appropriate for application to paper due to the large size of paper particles such as fibres. Similarly, Mie scattering is not applicable to the printed paper surface because neither the vehicle film nor the pigments appear as single particles of submicron size. Although the particle size of organic pigments is normally in the submicron range, nevertheless it is known that no dispersion method is capable of producing 100% monodispersed particles.

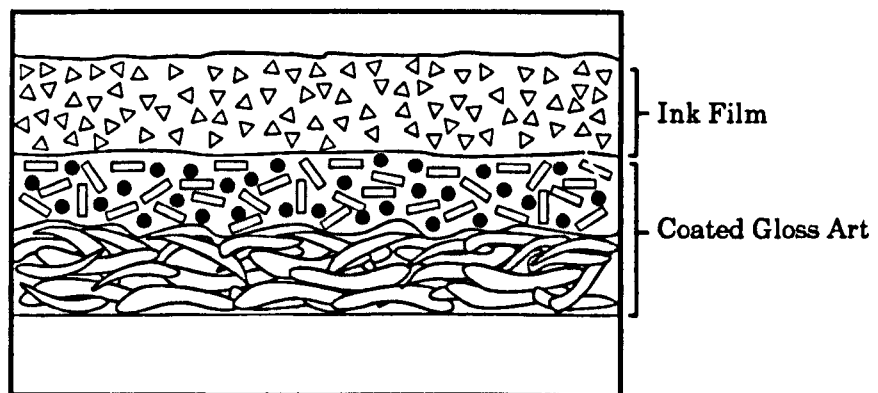
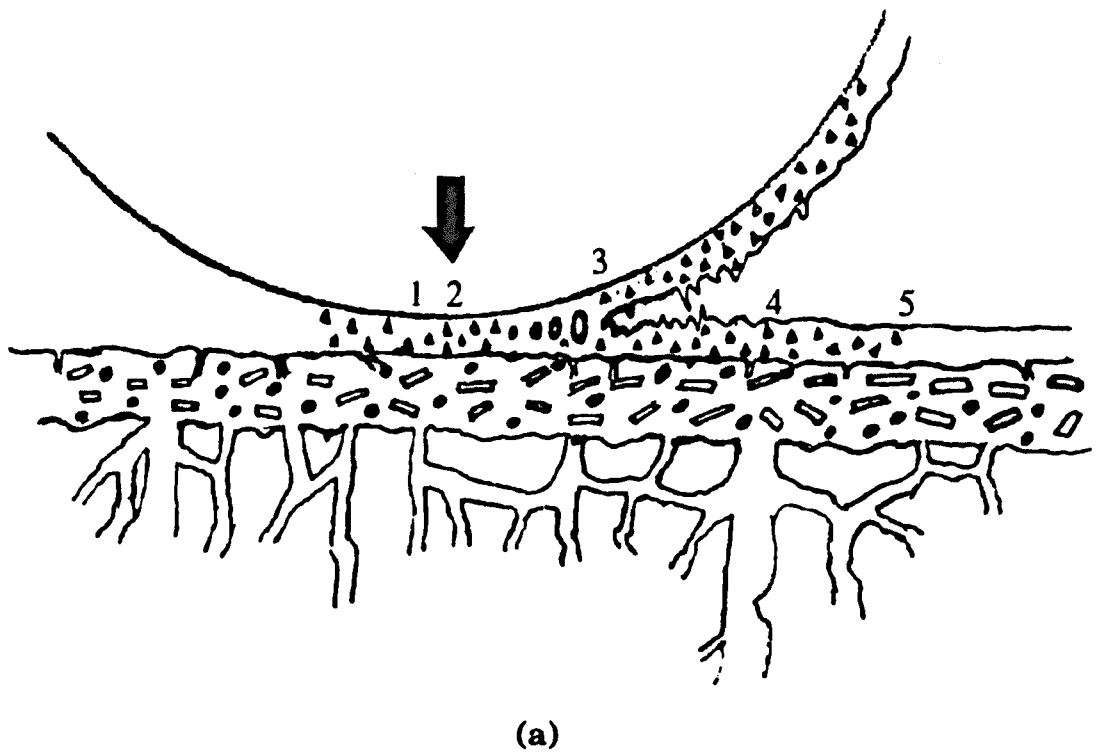


Leekly *et al.* [1970] observed that the reflectances of all printed surfaces were higher than those of unprinted surfaces due to the filling of paper crevices or pores and the leveling of the surface by the ink film. It was found from the present study that the reflectance of unprinted and printed surfaces for both gloss art and machine glazed does show a similar monotonic increase with increasing incident angle. However, the magnitude of the reflectances varied between printed and unprinted samples and not necessarily in the same direction as reported by Leekly *et al.* The reason for this argument is that the most influential factors which govern the magnitude of reflectance are the refractive index and the roughness of the surfaces. Regarding the refractive index of printed surfaces and paper surfaces, they are not significantly different as shown in the preceding section. Therefore, the dependency of the reflectance magnitude on the refractive index is of minor importance; it is considered that the surface roughness factor is predominant in this case. The results, on the other hand, agree well with Fetsko *et al.* [1973] who found that prints could be either smoother or rougher than the corresponding unprinted surface. ✕✕

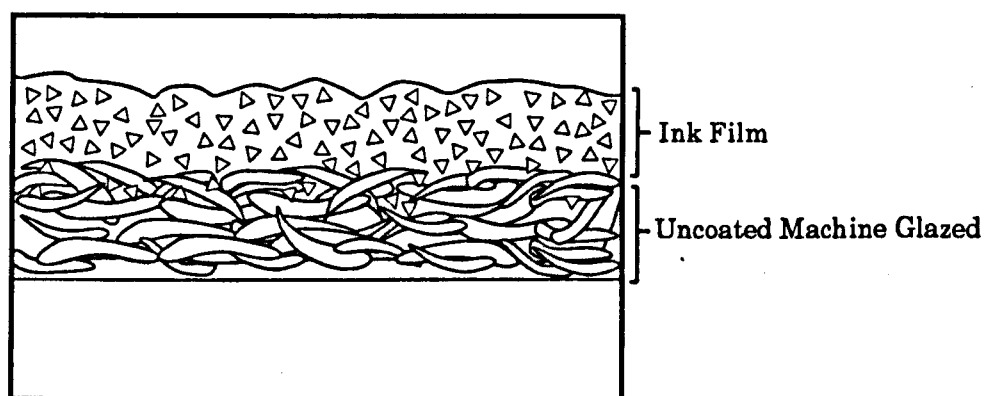
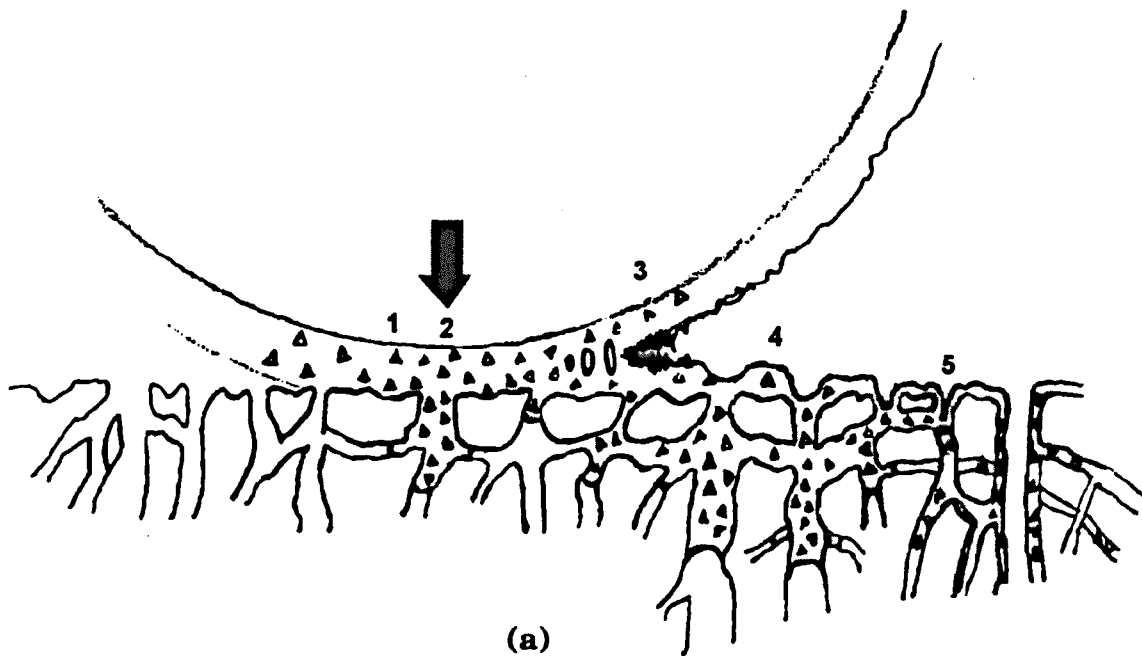
It is understood that a print is a result of ink and paper interactions in a printing process. Both ink transfer in the printing nip and ink drying after the nip are the crucial interactions for the formation of printed paper surfaces. Major stages of the development of the printed surface characteristics are illustrated in Figure 4.9a for gloss art prints and Figure 4.10a for machine glazed prints. Generally for both cases, the formation of a print occurs as follows. The ink first contacts the paper at the entrance of the printing nip (stage1). In the nip, the pressure applied not only compresses the porous structure of the paper but also hydraulically

impresses the ink into the compressed pores of the paper surface (stage 2). This is referred to as ink immobilization. At the nip exit, cavitation and filamentation occur in the ink film and eventually ink filaments split to give an amount of ink transfer on the surface (stage 3). After ink transfer, the paper decompresses and a redistribution of the ink may occur as some ink is imbibed into the expanded pores (stage 4) and this may continue to occur during the drying process. The ink film dries and results in a solid dried ink film on the surface (stage 5 and Figure 4.9b, 4.10b). It is known that ink penetration is generally involved in the drying of most printing inks.

The differences of paper properties between gloss art paper and machine glazed paper, as can be seen in Table 4.1, were found to affect these basic stages which in turn determine the resultant printed surface characteristics. The most important factors are smoothness, porosity and oil absorbency properties. Smoothness and compressibility of the paper determine the extent of ink contacting paper at the first stage. It is commonly known that a rougher paper is more compressible than a smooth paper. Gloss art paper, as a result of a coating process, shows a smoother surface and a less porous structure than machine glazed paper as indicated by the Parker Print-surf and the Bendsten porosity results. This leads to a better contact between the surface of gloss art and the ink. However, this effect of surface roughness on ink transfer in offset lithographic process is not significant due to the use of a resilient rubber blanket. Furthermore, the amount of ink transferred to machine glazed was found to be much greater (almost twice) than that transferred to gloss art even though the target print density of machine glazed is lower than that of gloss art, as shown in Table 4.4. This can be explained by the effects of both the surface roughness and the porous



**Figure 4.9 A schematic illustration of ink film formation for gloss art.**



**Figure 4.10 A schematic illustration of ink film formation for machine glazed.**

structure on ink immobilization of machine glazed paper. The results of ink transfer are in agreement with the study of DeGrâce & Dalphond [1989] who investigated the development of print density and print-through resulting from ink and paper interactions both during and after printing. They reported that the rougher the surface, the more ink is needed to obtain the desired optical coverage which in turn determines print density.

It is clearly seen that ink penetration plays an important role both in ink transfer and ink drying processes. Ink penetration in the nip is mainly governed by the printing pressure whereas ink penetration after the nip occurs by capillary action. It is believed that this phenomenon makes a dominant contribution to the characteristics of printed surfaces as will be further discussed. In studying penetration of ink into paper, the relationship between the pore sizes of paper and the pigment particle sizes of ink has to be taken into account.

In the case of gloss art paper, during impression where printing pressure is applied, only a small amount of ink could penetrate into the paper and thus fill in all the small surface pores. This is because of its relatively small pore size as compared to the pigment size. The average particle size of yellow pigments is approximately 0.2 microns [Vernardakis, 1984] whereas the pore size of gloss art [assumed from the study of Bristow & Berglenblad, 1992] may be approximately 0.1 microns. Moreover, it should be noted that when the pigments are dispersed into the ink vehicle system, some aggregates or flocculates may occur. After free ink film splitting, most of the ink including most pigments remain on the surface. An earlier study of Fetsko [1958] indicated that the magnitude of the free ink

l  
film splitting to coated paper is approximately 40% which is twice the magnitude of that to uncoated paper. After ink transfer, only the ink vehicle penetrates further into the paper as part of the ink drying process. Thus, there is a separation of ink vehicle from the pigment which is known as a filtration phenomenon [Coupe & Smith, 1956; Christensen, 1967; Lepoutre *et al.*, 1979]. However, vehicle penetration was also restricted by the porous structure itself. As the penetration slowly proceeds and the ink vehicle drains away from the pigments, ink vehicle capillaries are left between the pigment particles leading to gel characteristics of the ink which tends to retain the ink vehicle on the surface. Therefore, there is sufficient ink vehicle to provide a covering layer for pigment particles in the ink film to result in a uniform printed surface as shown schematically in Figure 4.9b. This ink vehicle film can be considered to correspond with Barkas' mirror facets. Printed gloss art samples when evaluated by the optical print smoothness method yielded macrosmoothness and microsmoothness values not significantly different from those for unprinted surfaces (see Table 4.7 and Figure 4.4). Therefore, it is concluded that printed gloss art is no less smooth than the unprinted surface. Regarding ink transfer parameters, it can be concluded that it is the ink film splitting factor and not the ink immobilization factor which significantly determines the characteristics of printed surfaces for coated papers. x

In the case of machine glazed, a large amount of ink including the pigments was impressed into the compressed pores of the paper surface during impression owing to its more porous structure. The pore sizes for uncoated papers are in the microns range without any dominant size, unlike coated papers [Banks, 1975; Bristow & Berglenblad, 1992]. This is because of

the tendency of the fibres to aggregate into large lumps during the manufacture of the paper. It can be seen from the Bendsten porosity results (Table 4.1) that the porosity of machine glazed is approximately 170 times greater than that of gloss art. Under the same printing conditions and ink film thickness on the blanket, the ink film splitting portion of machine glazed would be less than that of gloss art. After ink transfer, ink vehicle from the immobilization portion would tend to penetrate further into the smaller pores, leaving the large pores open. Some of the pigment particles may accompany the ink vehicle. Thus, the extent of filtration phenomena is not seen to be significant. Ink vehicle from the splitting portion on the surface also penetrates, but relatively slowly, into the body of the paper by diffusion or through small pores of the surface. As a consequence, the ink film appears to follow the contour of the unprinted surface (Figure 4.10b). The ink which remains on the surface would be expected to behave as highly inclined mirror facets within the macro regions. Printed machine glazed when evaluated by the optical method therefore yields a lower value in the macrosmoothness as compared to the unprinted surface. Regarding the ink transfer parameters, it can be concluded that ink immobilization plays a more important role in determining the characteristics of printed surfaces for uncoated papers.

In addition, it can be concluded that porosity is the most crucial factor in the formation of printed surface characteristics in this part of the study. The scanning electron micrographs of both unprinted paper surfaces (Figures 4.6-4.7) reveal the significant differences of the surface pore sizes of coated gloss art and uncoated machine glazed. The micrograph of gloss art printed with heatset yellow ink shows a distinct ink layer covering the paper. The micrographs of machine glazed printed with this ink do not show the

same details of ink layering as in the case of gloss art. Nevertheless, they do show ink layering with fibre related structure. Comparing the micrographs between gloss art prints and machine glazed prints, provides support for the importance of the filtration phenomenon on the formation of printed surface characteristics.

## **Part II(a) - The effects of emulsification on print smoothness.**

### **4.3.3 Results - Part II(a).**

This study was aimed at the offset lithographic process, the presence of fountain solution is an important factor in this printing process and its effects on print smoothness were, therefore, investigated. Different water contents, 10%, 20% and 30% in the 'emulsion inks' were prepared with the heatset yellow ink. Gloss art and machine glazed were then printed with these 'yellow emulsion inks' to obtain a target print density, that is  $D \sim 0.92$  for gloss art and  $D \sim 0.84$  for machine glazed. The optical smoothness of these prints was evaluated (Results are given in Appendix E, Table E.1-E.4). Statistical methods were employed to determine whether the optical smoothness results of unprinted and printed samples are significantly different.

Tables 4.8-4.9 and Figures 4.11-4.12 summarize the macrosmoothness and microsmoothness of both gloss art prints and machine glazed prints respectively. To consider changes in the print smoothness, a reference standard needs to be established. Both gloss art and machine glazed printed with heatset yellow ink (no emulsified fountain solution in the ink) were used as reference standards.



The scanning electron micrographs of four 'yellow' prints for gloss art and machine glazed samples are shown in serial order for comparison in Figure 4.13-4.14 repectively.

Table 4.8 Print smoothness summary: gloss art printed with heatset yellow ink and the 'yellow emulsion inks'. Each result given is the average value of twenty different points for each sample (See Appendix E).

(a) Macrosmoothness (Sm).

Sample	Sm ( $\bar{X}$ )	S.D.	95% C.I. for $\bar{X}$	Min.	Max.
GA + 100% Y- ink	0.974	0.084	0.935 to 1.014	0.855	1.133
GA + 90% Y- ink : 10% F.S.	1.023	0.107	0.973 to 1.073	0.885	1.210
GA + 80% Y- ink : 20% F.S.	1.010	0.090	0.967 to 1.052	0.853	1.225
GA + 70% Y- ink : 30% F.S. <sup>a</sup>	0.838	0.077	0.802 to 0.874	0.668	0.943

a - Sm was significantly different from the other three printed samples. (See Appendix F and Figure 4.11).

(b) Microsmoothness (Su).

Sample	Su ( $\bar{X}$ )	S.D.	95% C.I. for $\bar{X}$	Min.	Max.
GA + 100% Y- ink	0.932	0.052	0.908 to 0.957	0.785	0.995
GA + 90% Y- ink : 10% F.S.	0.931	0.043	0.911 to 0.951	0.858	0.991
GA + 80% Y- ink : 20% F.S.	0.932	0.042	0.912 to 0.951	0.855	0.983
GA + 70% Y- ink : 30% F.S.	0.911	0.054	0.885 to 0.936	0.774	0.996

No significant differences were observed.

Table 4.9 Print smoothness summary: machine glazed printed with heatset yellow ink and the 'yellow emulsion inks'. Each results given is the average value of twenty different points for each sample (See Appendix E).

(a) Macrosmoothness ( $S_m$ ).

Sample	$S_m (\bar{X})$	S.D.	95% C.I. for $\bar{X}$	Min.	Max.
MG + 100% Y- ink	0.089	0.015	0.082 to 0.096	0.071	0.126
MG + 90% Y- ink : 10% F.S.	0.087	0.020	0.078 to 0.096	0.061	0.121
MG + 80% Y- ink : 20% F.S. <sup>a</sup>	0.065	0.012	0.060 to 0.070	0.037	0.088
MG + 70% Y- ink : 30% F.S. <sup>a</sup>	0.065	0.011	0.060 to 0.070	0.044	0.087

a -  $S_m$  were significantly different from machine glazed printed with 100% heatset yellow ink and 10% water content 'yellow emulsion ink' (See Appendix F and Figure 4.12).

(b) Microsmoothness ( $S_u$ ).

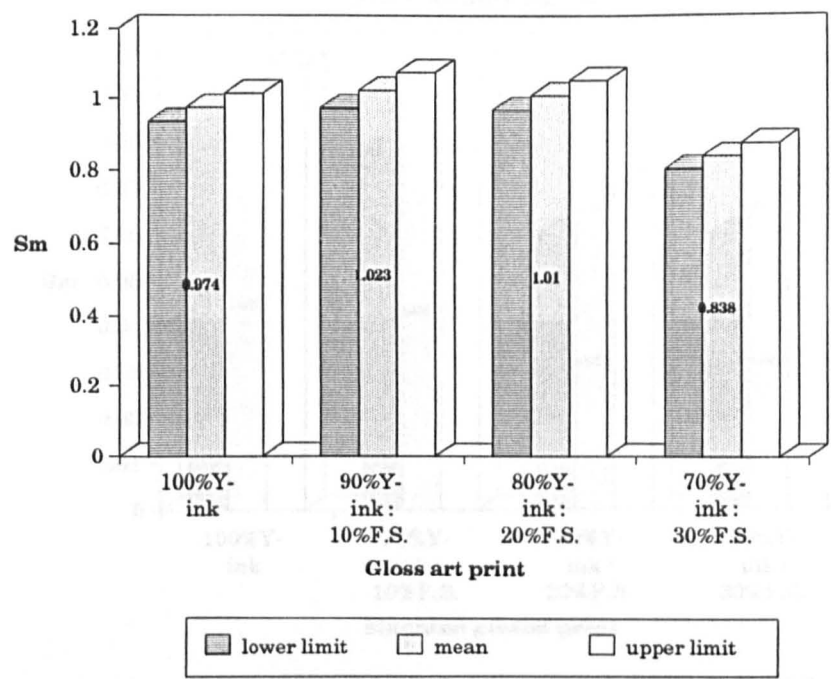
Sample	$S_u (\bar{X})$	S.D.	95% C.I. for $\bar{X}$	Min.	Max.
MG + 100% Y- ink	0.874	0.056	0.848 to 0.901	0.750	0.958
MG + 90% Y- ink : 10% F.S.	0.855	0.101	0.808 to 0.902	0.667	0.973
MG + 80% Y- ink : 20% F.S.	0.849	0.094	0.805 to 0.893	0.703	0.998
MG + 70% Y- ink : 30% F.S.	0.846	0.076	0.810 to 0.881	0.696	0.968

No significant differences were observed.

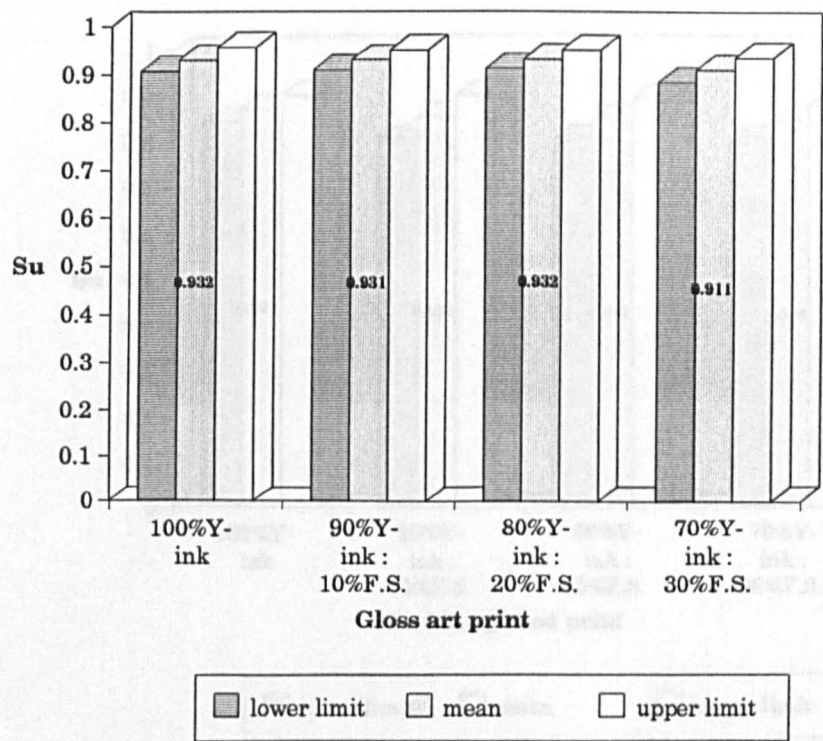
Figure 4.11 Print smoothness of gloss art (pigmented prints).

(a) Macrosmoothness.

(b) Microsmoothness.

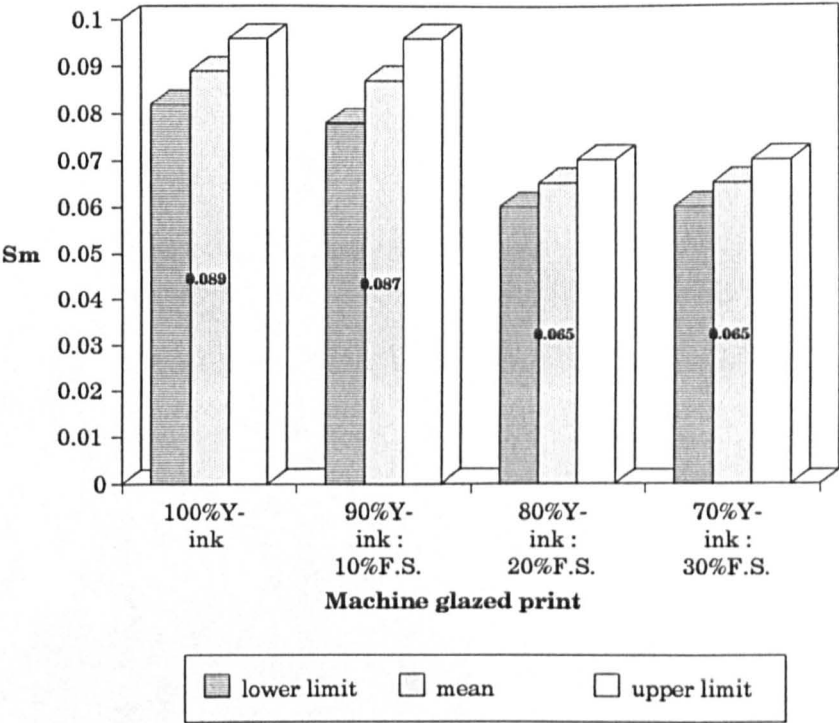


(a)

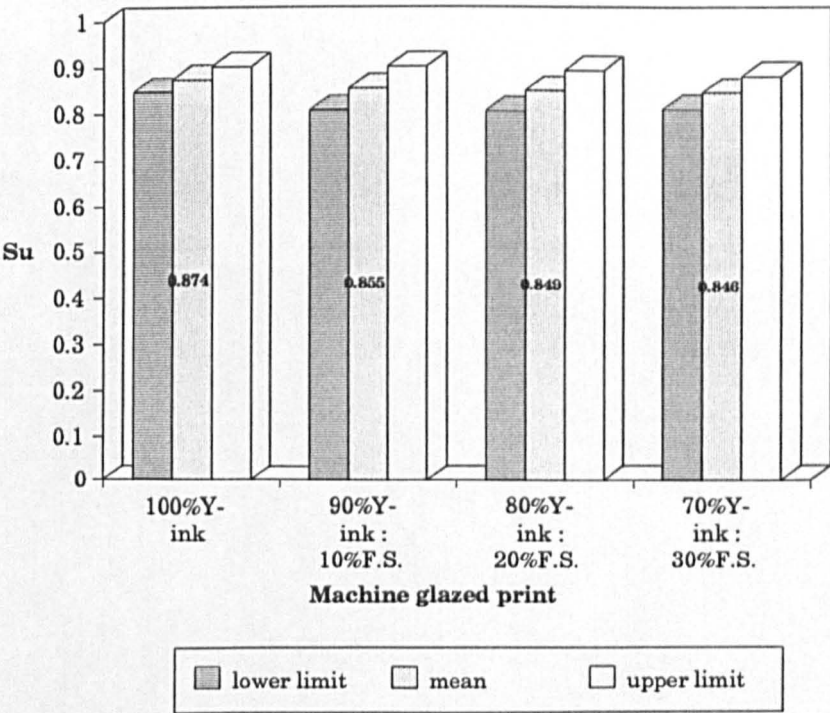


(b)

Figure 4.12 Print smoothness of machine glazed (pigmented ink).  
 (a) Macrosmoothness.  
 (b) Microsmoothness.

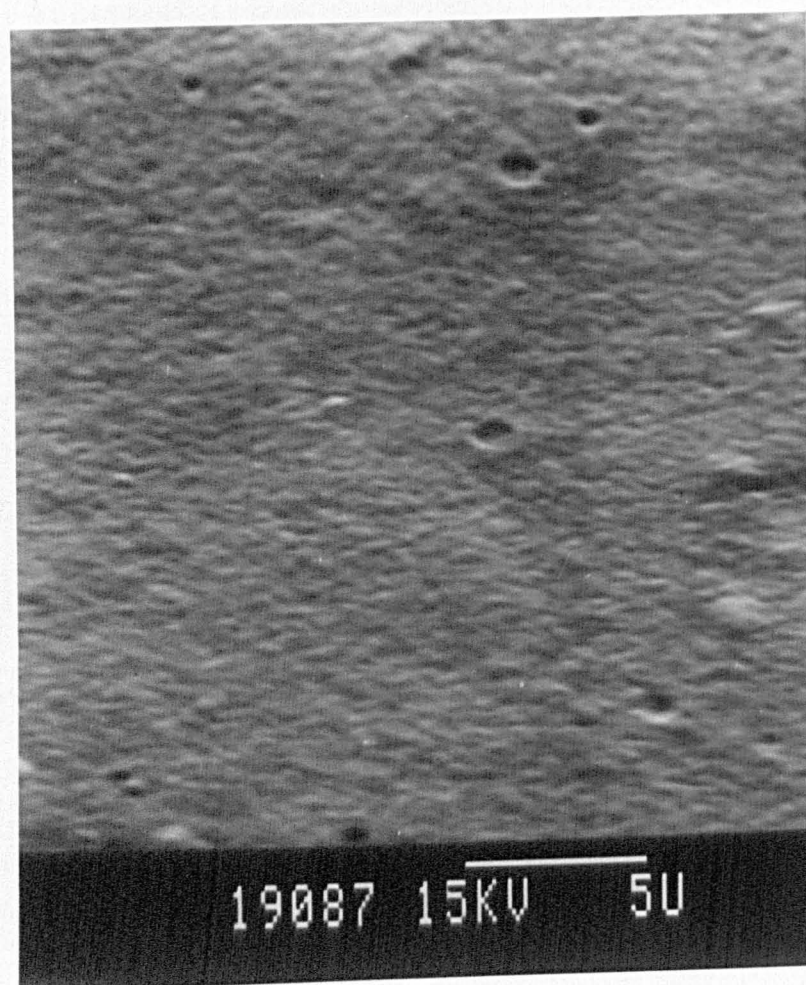


(a)

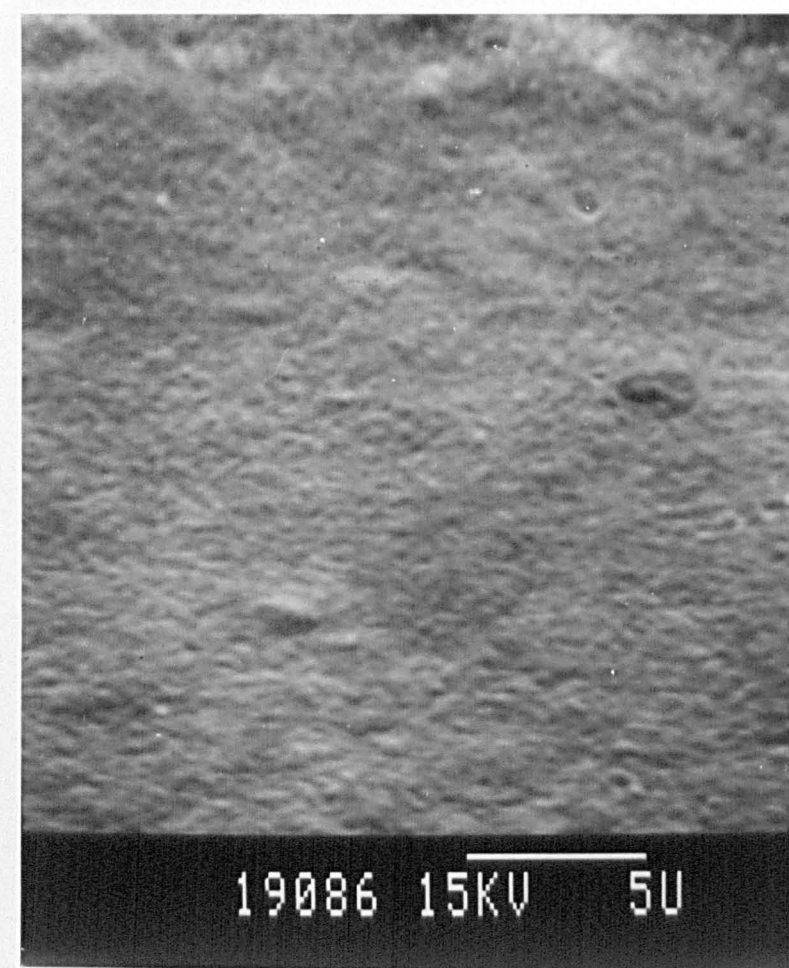


(b)

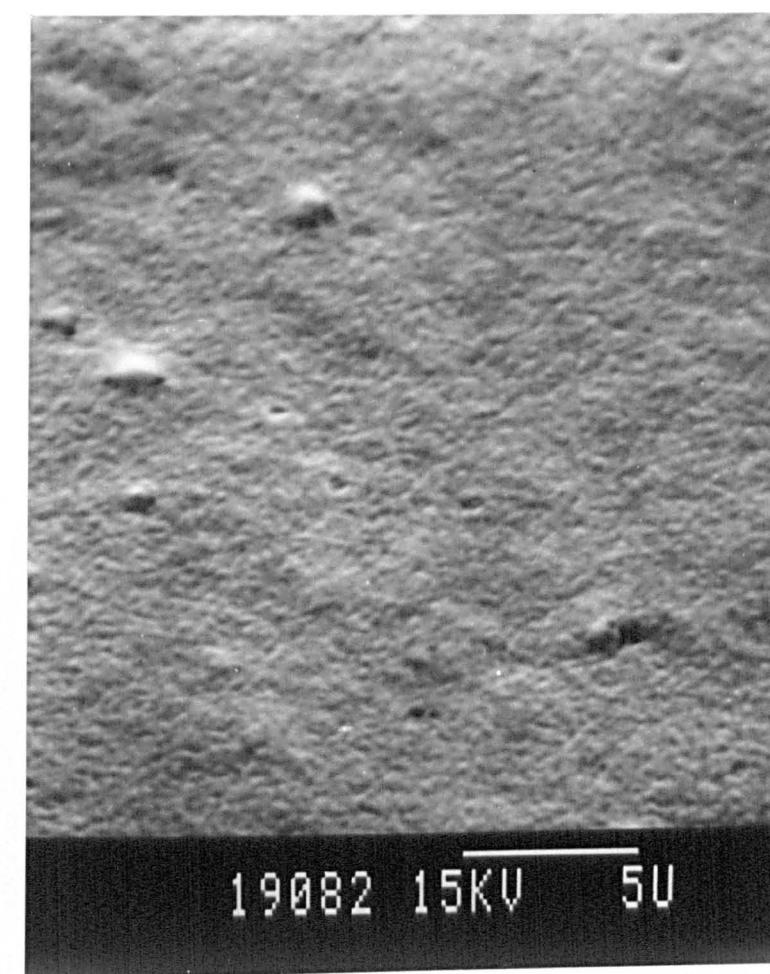




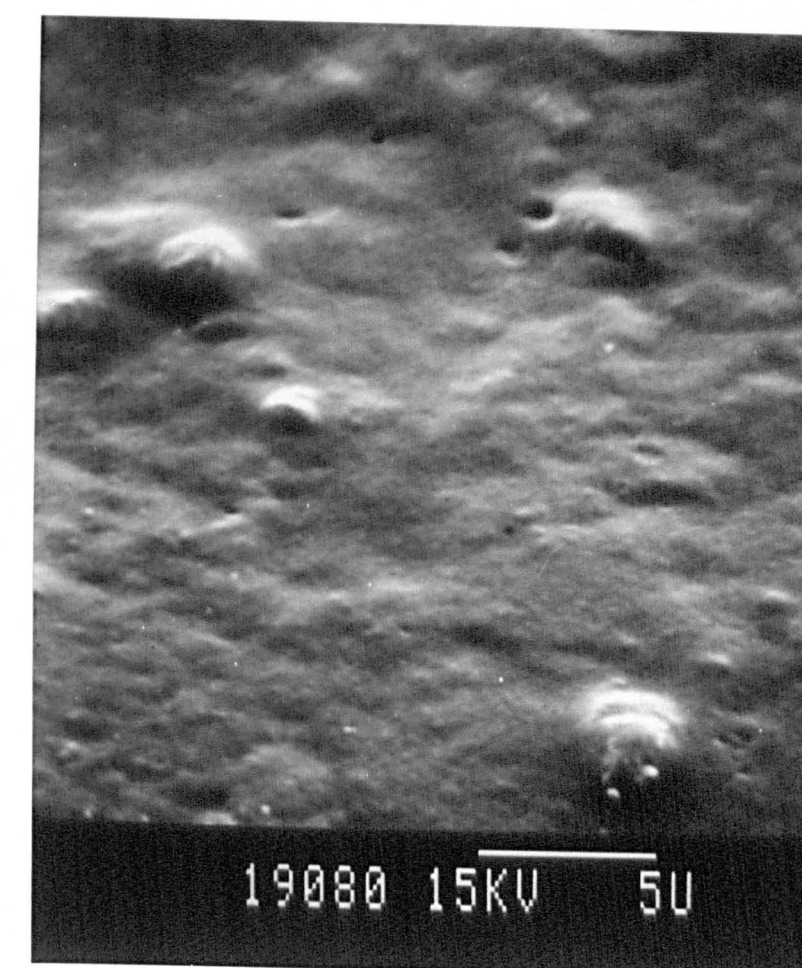
(a)



(b)



(c)



(d)

**Figure 4.13 Scanning electron micrographs x 5000.**

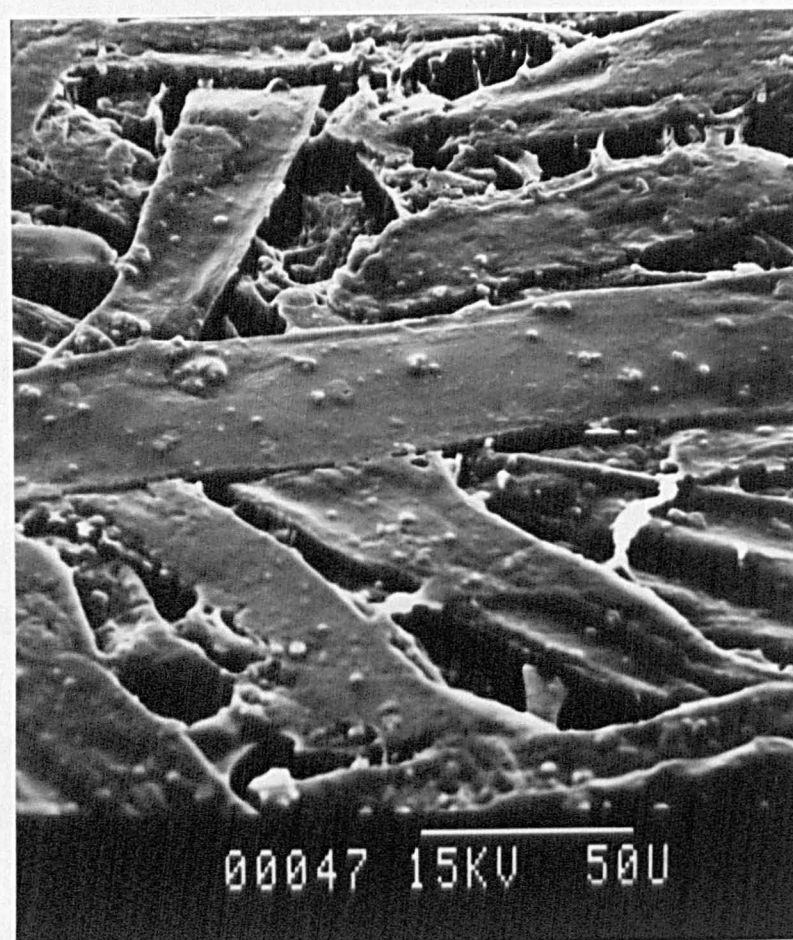
(a) GA printed with 100%Y-ink.

(b) GA printed with 90%Y-ink:10%F.S.

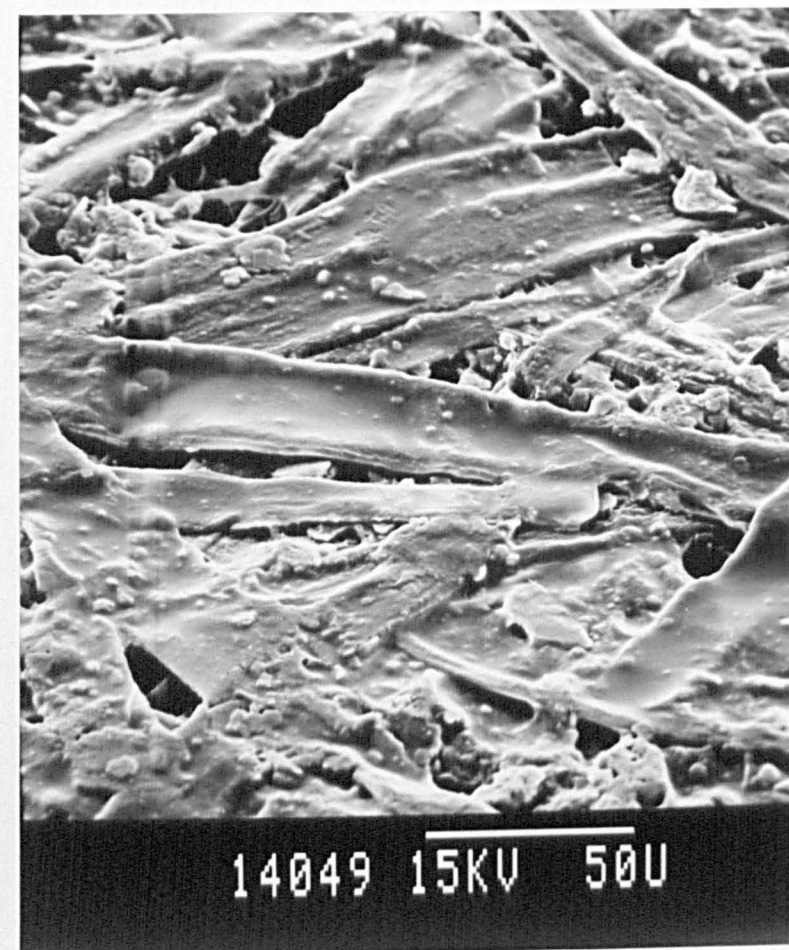
(c) GA printed with 80%Y-ink:20%F.S.

(d) GA printed with 70%Y-ink:30%F.S.

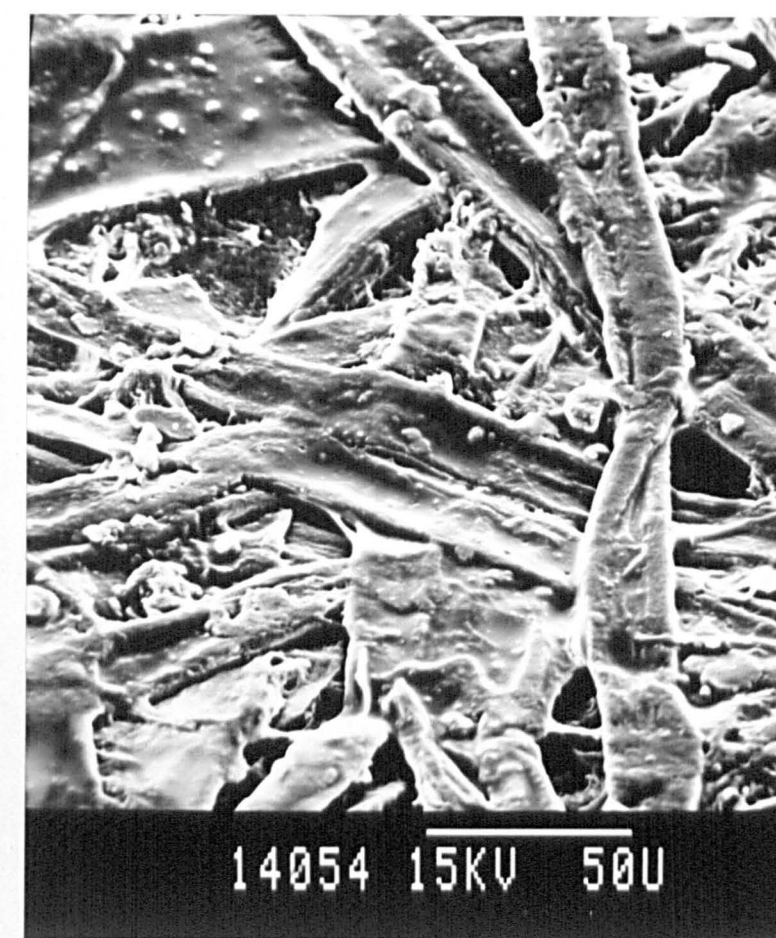




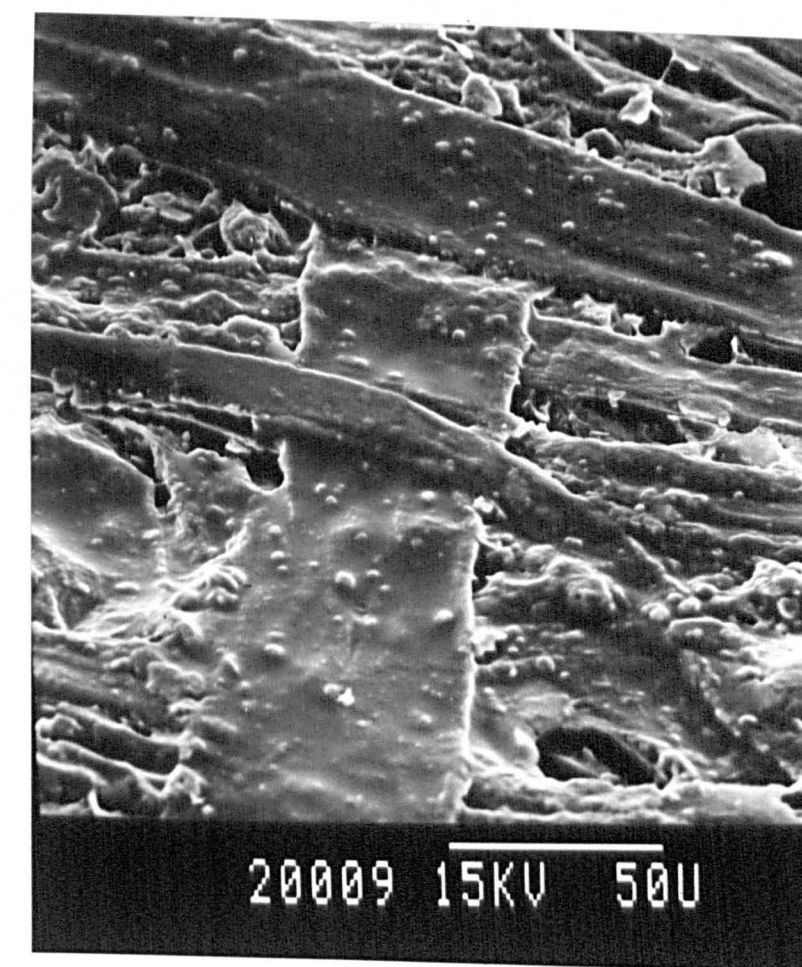
(a)



(b)



(c)



(d)

**Figure 4.14 Scanning electron micrographs x 600.**

- (a) MG printed with 100%Y-ink.**
- (b) MG printed with 90%Y-ink:10%F.S.**
- (c) MG printed with 80%Y-ink:20%F.S.**
- (d) MG printed with 70%Y-ink:30%F.S.**

The analysis of variance and the multiple comparison test (See Appendix F), in the case of gloss art prints, revealed that only the macrosmoothness mean value of gloss art printed with 30% water content 'yellow emulsion ink' was significantly different from those of the other three printed samples. No significant differences were observed for the macrosmoothness of these prints. The macrosmoothness of printed gloss art decreased from a value of 0.974 (printed with no fountain solution emulsified in the ink) to a value of 0.838 (printed with 30% water content emulsified in the ink). The printed surface of gloss art printed with 30% water content 'yellow emulsion ink' is therefore a rougher surface.

In the case of machine glazed, these statistical methods revealed that the macrosmoothness mean values of machine glazed printed with 20% and 30% water content 'yellow emulsion inks' were significantly different from those printed with heatset yellow ink and 10% water content 'yellow emulsion ink'. No significant differences were observed for the macrosmoothness of these prints.

It is interesting to note that within the range explored of emulsification, the macrosmoothness values of printed machine glazed can be characterized by two smoothness levels. One level included: the value of 0.089 from machine glazed printed with heatset yellow ink, which was used as a reference; and of 0.087 from machine glazed printed with 10% water content 'yellow emulsion ink'. The other level included machine glazed printed with 20% and 30% water content 'yellow emulsion inks', these resulted in the same lower macrosmoothness value of 0.065 and therefore fall into a rougher level.



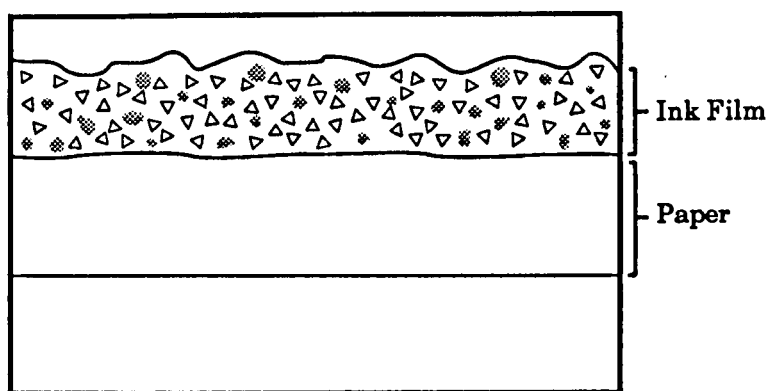
#### 4.3.4 Discussion - Part II(a).

There was a difference between the gloss art and the machine glazed samples; namely in the critical amount of fountain solution emulsified in the ink which would affect print smoothness. Nevertheless both show the same trend of a decrease in print smoothness with increasing amount of emulsified fountain solution. These printed surfaces are rougher. It is apparent that another important factor involved is the 'ink'. Therefore, in studying the effects of emulsification on print smoothness, it is necessary to consider the quality of these 'emulsion inks'. Table 4.5 gives the rheological properties of the 'yellow emulsion inks'. It can be seen that as the amount of emulsified fountain solution increases, the viscosity decreases. In this study, it also appears that tack decreases as the amount of emulsified fountain solution increases. The rheological properties of emulsion inks have been examined by many researchers and these findings are in agreement with those observed by Lavelle *et al.*, [1969] and Bassemir & Shubert [1985]. Other important factors are the size and the distribution of the emulsified droplets. It is believed that increasing the amount of emulsified fountain solution leads to the occurrence of more large droplets due to the greater probability of movements of small droplets which coalesce into one large droplet. This instability of emulsion ink was noted by Banks [1970]. In addition, it will be assumed that the technique used to prepare the 'yellow emulsion inks' is equivalent to that of Tasker *et al.*, [1983] who employed a three-roll lithobreak tester operating at a low speed of 100 rpm and found that the droplet sizes never exceeded 2 microns with a *good* heatset ink. They also reported that inks which formed a wide range of emulsified droplet sizes above 8 microns upto 20 microns were problem inks. Also it is known that fountain solution having isopropanol produces emulsions of finer and more uniform droplets than that without

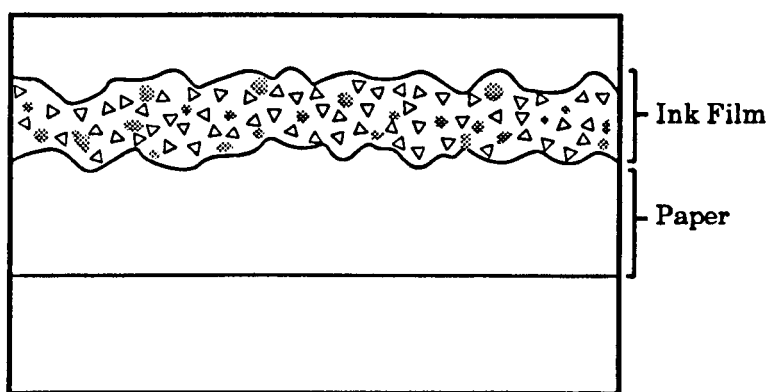
isopropanol [Braun, 1985]. Therefore, it will be assumed that at low amount of water content in this 'yellow emulsion ink', the average droplet sizes are in a range of less than 2 microns. When the emulsified amounts exceed the critical amount, the sizes of the emulsified droplets vary considerably.

As discussed in Part 1, both ink transfer and ink drying processes determine the characteristics of printed surfaces. Therefore, it is postulated that the roughness of these printed surfaces arose from interactions between the 'emulsion inks' and the papers; where the relevant phenomena are as discussed in the following. Figure 4.15 is a schematic illustration of three phenomena which are postulated to be the causes of print roughness.

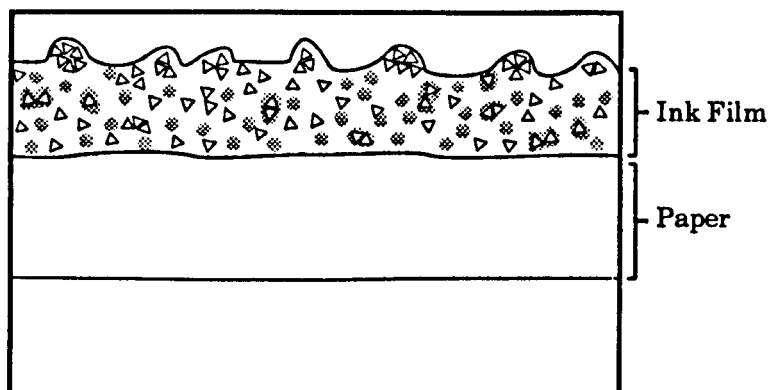
Considering the ink transfer phenomena. It is accepted that the extent of ink contact with paper depends mostly on the paper properties. The extent of contact also depends to a lesser extent on ink properties. It was reported that short inks produce less contact between ink and paper [Walker & Fetsko, 1955]. Short ink generally has higher tack. The tack of 'emulsion inks' depends on the amount of emulsified fountain solution; and as found in the present study, the tack decreases with 'emulsion inks'. Therefore, it can be assumed that with 'emulsion inks', the ink/paper contact should be more. However, as shown in Table 4.5, the decrease in tack of 'yellow emulsion inks' is only 7% whereas the decrease in viscosity is more than 60%. In addition to the use of a resilient rubber blanket, it is considered that the influence of ink tack on ink/paper contact in these cases can be negligible.



(a)



(b)



(c)

**Figure 4.15 A schematic illustration of rough printed surfaces caused by**  
**(a) Cavitation and filamentation.**  
**(b) Ink penetration.**  
**(c) Pigment aggregation.**

Note:  $\triangle$  --- yellow pigment

$\odot$  --- emulsified fountain solution

Ink immobilization refers to the ink which was forced to penetrate into the paper by printing pressure. It was found that with decreasing ink viscosity, ink immobilization increased [Schaeffer, et al., 1961]. Accordingly, it is believed that more ink was impressed into the paper with 'emulsion inks' according to the amount of fountain solution emulsified.

1. Cavitation and filamentation (Figure 4.15a). At the nip exit, cavitation and filamentation occur. These phenomena are related to ink film splitting. It was found that pigments in an oil vehicle behave as foreign nuclei in the dispersion system and enhance cavities formation [Myer et al., 1959]. From this knowledge, it is postulated that the presence of emulsified droplets in the ink film may also act as nuclei for cavitation and filamentation. However, as a result of increasing the amount of emulsified fountain solution, the droplet sizes are in a wider range (possibly up to 20 microns) and more large droplets can occur. These droplets initiate cavities at different positions within the ink film; large droplets initiated cavitation more readily than small droplets [Lyne & Aspler, 1982]. As a consequence, at the moment of ink film splitting, the ink filaments rupture unevenly. After ink film splitting, the smoothness of the printed surface strongly depends on the flow property of the transferred ink film, to level out evenly before the ink sets. This explanation is not completely contrary to Surland [1983] who suggested a 'cubic film splitting model' to explain the effect of emulsification on ink film splitting in the ink transfer process. Although he concluded that emulsion inks transfer more evenly than the corresponding inks, it has to be commented that the sizes of the disruptive particles should necessarily be considered. Cavitation and filamentation are postulated to be a cause of the roughness of printed surfaces. Many previous studies support this. For

example, it has been reported that uniform small droplets give the best gloss and uniform large droplets can give a better gloss than uneven-sized droplets [Dahms & Hafner, 1988]. Low water pickup inks were found to produce better prints than high water pickup inks; with reference to the print qualities studied namely print density, print gloss, dot gain, trapping and print mottle [Bassemir *et al.*, NPIRI 1990].

The critical point for the amount of emulsified fountain solution which causes rough printed surfaces, differs between gloss art and machine glazed; and this can be understood in terms of cavitation and filamentation as follows. In the case of gloss art, it has been shown that very little ink penetrates into the paper, most of the ink remains on the surface. Thus, it is the presence of those emulsified fountain solution droplets, within the ink layer on the surface, which plays an important role in cavitation and filamentation; and the roughness effect on the printed surface was found at 30% water content only. In the case of machine glazed, although more ink has penetrated during impression, nevertheless the amount of ink transferred was not sufficient to fill up the large pores. Air released from the large pores behaves in analogous manner to large emulsified droplets. Thus, at 20% water content, both emulsified fountain solution droplets within the ink layer on the surface and air from the paper surface initiate cavitation.

2. Ink penetration (Figure 4.15b). As previously discussed, it was found that the viscosity decreases after increasing the amount of emulsified fountain solution. It is also postulated that more large droplets occur with increasing the amount of emulsified fountain solution. These two factors integrate in the penetration process to cause the printed surfaces to become

rough. In the case of gloss art, owing to the lower viscosity of the 30% water content 'yellow emulsion ink', more ink can penetrate in the nip; and after the nip the ink vehicle penetrates further. Because of a wide range of sizes of emulsified droplets within this emulsion ink, the coating layer or both the coating layer and the fibres underneath become affected by this emulsion ink, more at some points than at others. This ink penetration can cause release of internal stress of the coating layer, fibre swelling, and bond breakage; leaving unbalanced stresses. Both the coating layer and the fibres underneath have been displaced; leading, at particular points, to localized relative displacements. As the ink film is dried on this surface, a rough printed surface is formed. The explanation in the case of machine glazed prints also follows a similar pattern although the critical point is at 20% water content for machine glazed. The difference in the critical point is due to the rougher and more porous structure of machine glazed than gloss art. Thus, at 20% of emulsified fountain solution in the yellow ink, there is a sufficient penetration depth (due to lowered viscosity) to cause disruption of the fibre network geometry of machine glazed. Swelling and debonding further increase the paper roughness and surface porosity and thus ink vehicle and pigments may penetrate further into deeper pores where they do not contribute to smoothness. This phenomenon is postulated to be analogous to water and paper interaction during paper coating [Skowronski & Lepoutre, 1985].

3. Pigment aggregation (Figure 4.15c). It was found that pigments show a preference for water rather than the vehicles and all pigment systems emulsified greater amounts of water than did the corresponding vehicles [Lavelle *et al.*, 1969; Banks, 1970; Braun, 1985]. From these findings, it can

be envisaged that the emulsified fountain solution may preferentially spread as a thin film around the pigment surfaces. With increasing amounts of emulsified fountain solution, more pigment is covered with a layer of fountain solution film. Further ink penetration, after ink transfer by capillary action, appears to be affected by the penetration of ink in the nip. As previously discussed, ink penetration in the nip can cause fibre swelling which causes a reduction in pore size. It is known that narrower pores exert a higher capillary negative pressure and that the rate of penetration is dependent on the viscosity. Therefore, more ink vehicle of 30% water content 'yellow emulsion ink' can penetrate after ink transfer; this can lead to a redistribution of components within the ink layer on the surface, which causes the pigments to join together with the coalescence of the fountain solution film. The effect of pigment aggregates on the roughness of printed surfaces will be seen only if there is not sufficient ink vehicle to form film thick enough to cover these pigment aggregates. The retention of ink vehicle within the ink film is a result of vehicle capillaries formed within the ink film during the drying process. This further indicates an important role of the pigment in the ink. It is reasonable to conclude that this effect of pigment aggregation should be clearly seen in the case of gloss art prints because more pigments remain on the surface than on machine glazed prints. .

The micrographs of all 'yellow' prints except that printed with 30% water content 'yellow emulsion ink' show similar printed surface characteristics. The micrograph of gloss art printed with 30% water content 'yellow emulsion ink' illustrates that some areas protruded from the ink film. This is believed to be the yellow pigment aggregates. Further work is required to confirm whether the protruding elements are due to localized

coating or fibre rising or to pigment aggregates remaining on the uppermost layer.

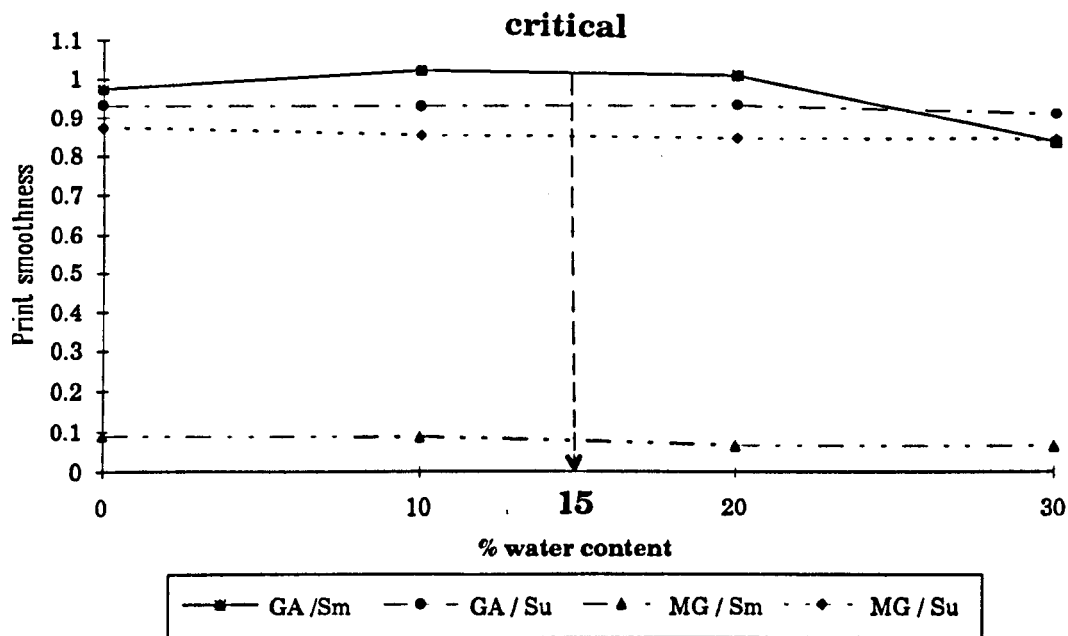
In comparisons between the micrographs of four 'yellow' machine glazed prints, it can be seen that the printed surface characteristics of machine glazed printed with 20% and 30% water content 'yellow emulsion ink' are slightly different from the other samples. That is the fibre structures shown for these two samples appear to be less compact and raised.

### **Critical water content.**

Rosenberg [1986] has defined two types of water taken up by the ink in the offset lithographic process: emulsified water and surface free water; and suggested that a critical water content of every ink should be indicated, below which the free water was emulsified rapidly and is finally unable to keep the non image areas clean. Similarly, the present study considers the critical water content as the limit of emulsification above which print smoothness cannot be achieved. Figure 4.16 is the plot of print smoothness as a function of % water content. The results suggest that there is a trend for print smoothness to decrease from a point where the water content in the 'emulsion ink' is greater than 20% for gloss art print and 10% for machine glazed print. The interval of %water content emulsified in the ink under investigation was too wide. The effects of emulsification within the range of 10%-20% appear to be interesting points if for example the same job is aimed to be printed on different papers under the same printing conditions. However, it may be deduced from Figure 4.16 that the critical amount of fountain solution which can be emulsified in this heatset yellow ink is 15% water content. This, in fact, suggests that if ink and water balance in the



press is set so that this heatset yellow ink emulsifies approximately 15% fountain solution, good quality can be expected for the resulting prints. Huelsman [1952] stated that the inks which have too low water absorption are bad, inks which absorb 15-20% of water are good and inks which absorb over 25% of water are definitely bad.



**Figure 4.16 Print smoothness as a function of % water content.**

The results from this experimental part lead to another hypothesis that pigment aggregation may play a more important role in decreasing print smoothness. Further experimental studies were therefore undertaken to examine the role of pigments in the emulsification and the effects on print smoothness.

## **Part II(b) - Study of the role of the pigments in the 'emulsion inks' and the effect on print smoothness.**

### **Experimental methods.**

To examine this effect, this part of the experiment was designed to use a specially made unpigmented ink. This is an 'ink' having the same formulation as the heatset yellow ink with the yellow pigments excluded and was prepared and supplied by Coates Lorilleux International. This will be referred to as U-ink. Experimental methods used were repeated as described in the previous part; with some minor differences which will be noted.

1. Pre-emulsified unpigmented inks. It was found that the print smoothness values of both gloss art and machine glazed decreased when printed with 30% water content 'yellow emulsion ink', therefore 30% water content 'unpigmented emulsion ink' was prepared to investigate the effect.

2. Printing. It is not possible to use density readings to control the test prints when printing with an unpigmented ink. Therefore, the amounts to be transferred of unpigmented ink and 30% water content 'unpigmented emulsion ink' were calculated; so as to be equivalent to the corresponding amounts of heatset yellow ink, (excluding the yellow pigments), in the previous print-run. These amounts of ink transfer are given in Table 4.10.

Table 4.10 The amounts of ink transfer for gloss art and machine glazed printed with 'unpigmented inks'.

Sample	ink transfer (g/m <sup>2</sup> )
GA + 100% U - ink	0.67
GA + 70% U - ink : 30% F.S.	0.77
MG + 100% U - ink	1.36
MG + 70% U - ink : 30% F.S.	1.41

3. Drying as previously described.

4. The rheological properties of the unpigmented ink and the corresponding 30% water content 'emulsion ink' were determined and are given in Table 4.11. It can be seen that the viscosity of this unpigmented ink is lower whereas the tack is greater than that of heatset yellow ink (Table 4.5). Nevertheless, both viscosity and tack decreased when fountain solution was emulsified in this unpigmented ink.

Table 4.11 The rheological properties of unpigmented ink and the corresponding 'emulsion ink'.

Property	Ink sample	
	100 % U - ink	70% U - ink + 30% F.S.
Viscosity, (Poise) 32°C	49	31
Tack, ('Tack-o-Scope' unit), 25°C	138	119

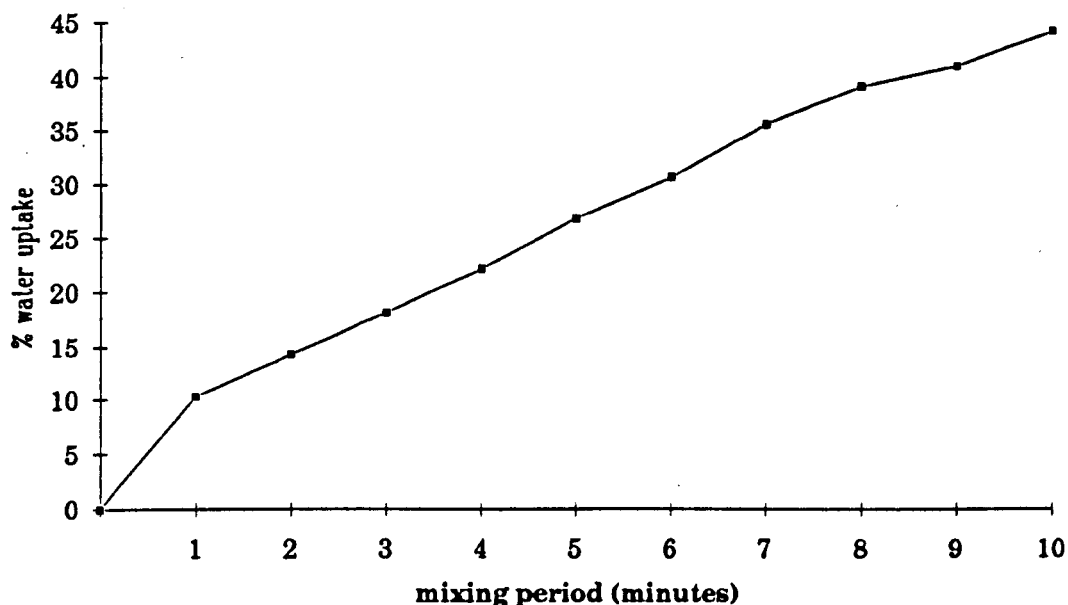
5. Print smoothness and print gloss were evaluated. Print smoothness results are given in Appendix E (Tables E.5-E.8). Print gloss results of both gloss art and machine glazed printed with unpigmented ink and 'unpigmented emulsion ink' are presented in Table 4.12. It can be noted that the print gloss values between each pair of 'unpigmented' prints for both gloss art and machine glazed samples are not significantly different. However, comparing the corresponding samples in Table 4.6, significant differences also appear for the pigmented samples. Print gloss values of all gloss art prints are different from paper gloss of the unprinted samples. In the case of machine glazed samples, the gloss values for both paper and prints appeared to be so low that it may be assumed that the geometry of this glossmeter is not appropriate to determine such low gloss surfaces.

Table 4.12 Print gloss of gloss art and machine glazed printed with 'unpigmented inks'.

Sample	Gloss (%)
GA + 100% U - ink	67.3
GA + 70% U - ink : 30% F.S.	66.9
MG + 100% U - ink	9.8
MG + 70% U - ink : 30% F.S.	9.6

6. Statistical methods were applied to these optical smoothness results. Summaries of these statistical analyses are shown in Appendix F (Table F.6-F.9).

7. Water pickup test of this unpigmented ink was carried out. A graph plotted between % water pickup versus time is shown in Figure 4.17.



**Figure 4.17 The percentage water uptake of unpigmented ink.**

As compared to the percentage water uptake of heatset yellow ink in Figure 4.2, it can be seen that the amount taken up by the heatset yellow ink was almost twice the amount taken up by the unpigmented ink. This is in agreement with Chambers [1993] who used other pigmented and unpigmented systems and similar results were obtained. These results indicate a significant role of pigments in the mechanism of water uptake.

## **The role of pigment in the emulsification and its effect on print smoothness.**

### **4.3.5 Results - Part II(b)**

Tables 4.13-4.14 and Figures 4.18-4.19 summarize macrosmoothness and microsmoothness of four printed samples, namely: prints printed with heatset yellow ink, printed with 30% water content 'yellow emulsion ink', printed with unpigmented ink and printed with 30% water content 'unpigmented emulsion ink'; for both gloss art and machine glazed.

Scanning electron micrographs of unprinted sample, and samples printed with unpigmented ink and printed with 30% water content 'unpigmented emulsion ink' for gloss art and machine glazed papers are shown in Figures 4.20-4.21 respectively.

Considering the results from the analysis of variance and multiple comparison tests, in the case of these printed gloss art samples (Table 4.13 and Figure 4.18), these statistical methods bring out some additional interesting information relating to pigments. No significant difference was observed for gloss art prints between the macrosmoothness mean values for unpigmented ink with and without 30% water content; but both were significantly different from those for gloss art printed with heatset yellow ink and with 30% water content 'yellow emulsion ink'. No significant differences were observed in the microsmoothness between these prints. It can be concluded that both 'unpigmented' prints have smoother surfaces; and gloss art printed with 30% water content 'yellow emulsion ink' possesses the roughest printed surface characteristics of the group.

In the case of the printed machine glazed samples (Table 4.14 and Figure 4.19), the statistical methods revealed that the macrosmoothness mean value, using unpigmented ink, was significantly different from that for 30% water content 'unpigmented emulsion ink'. Moreover, both were significantly different from those for heatset yellow ink and for 30% water content 'yellow emulsion ink'. No significant differences were observed, however, in the microsmoothness between these prints. It should be noted that within the machine glazed prints, the smoothest printed surface was obtained when printed with 30% water content 'unpigmented emulsion ink' whereas the roughest printed surfaces were obtained when printed with the amount of 20% (or more) water content in 'yellow emulsion ink'.

Table 4.13 Print smoothness summary: gloss art printed with heatset yellow ink, unpigmented ink and their corresponding 30% water content 'emulsion inks'. Each result given is the average value of twenty different points for each sample (see Appendix E).

(a) Macrosmoothness ( $S_m$ ).

Sample	$S_m (\bar{X})$	S.D.	95% C.I. for $\bar{X}$	Min.	Max.
GA + 100% Y-ink	0.974	0.084	0.935 to 1.014	0.855	1.133
GA + 70% Y-ink : 30% F.S. <sup>a</sup>	0.838	0.077	0.802 to 0.874	0.668	0.943
GA + 100% U-ink <sup>b</sup>	1.130	0.125	1.072 to 1.189	0.914	1.363
GA + 70% U-ink : 30% F.S. <sup>b</sup>	1.172	0.123	1.115 to 1.230	0.946	1.377

a -  $S_m$  was significantly different from the other three prints (See Appendix F and Figure 4.18).

b -  $S_m$  was significantly different from gloss art printed with heatset yellow ink (See Appendix F and Figure 4.18).

(b) Microsmoothness ( $S_u$ ).

Sample	$S_u (\bar{X})$	S.D.	95% C.I. for $\bar{X}$	Min.	Max.
GA + 100% Y-ink	0.932	0.052	0.908 to 0.957	0.785	0.995
GA + 70% Y-ink : 30% F.S.	0.911	0.054	0.885 to 0.936	0.774	0.996
GA + 100% U-ink	0.945	0.035	0.928 to 0.961	0.861	0.994
GA + 70% U-ink : 30% F.S.	0.940	0.029	0.926 to 0.953	0.893	0.994

No significant differences were observed.



Table 4.14 Print smoothness summary: machine glazed printed with heatset yellow ink, unpigmented ink and their corresponding 30% water content 'emulsion inks'. Each result given is the average value of twenty different points for each sample (see Appendix E).

(a) Macrosmoothness (Sm).

Sample	Sm ( $\bar{X}$ )	S.D.	95% C.I. for $\bar{X}$	Min.	Max.
MG + 100% Y-ink	0.089	0.015	0.082 to 0.096	0.071	0.126
MG + 70% Y-ink : 30% F.S. <sup>a</sup>	0.065	0.011	0.060 to 0.070	0.044	0.087
MG + 100% U-ink <sup>b,c</sup>	0.104	0.013	0.098 to 0.110	0.086	0.132
MG + 70% U-ink : 30% F.S. <sup>c</sup>	0.116	0.026	0.104 to 0.128	0.083	0.169

a - Sm was significantly different from the other three printed samples. (see Appendix F and Figure 4.19).

b - Sm was significantly different from machine glazed printed with 30% water content 'unpigmented emulsion ink' (see Appendix F and Figure 4.19).

c - Sm was significantly different from machine glazed printed with heatset yellow ink (see Appendix F and Figure 4.19).

(b) Microsmoothness (Su).

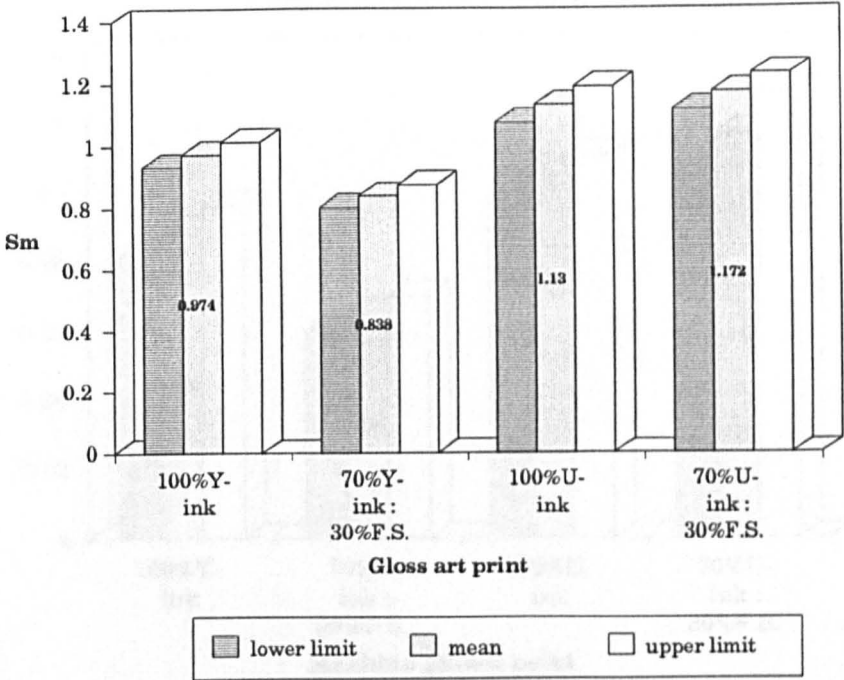
Sample	Su ( $\bar{X}$ )	S.D.	95% C.I. for $\bar{X}$	Min.	Max.
MG + 100% Y-ink	0.874	0.056	0.848 to 0.901	0.750	0.958
MG + 70% Y-ink : 30% F.S.	0.846	0.076	0.810 to 0.881	0.696	0.968
MG + 100% U-ink	0.854	0.092	0.811 to 0.897	0.702	0.987
MG + 70% U-ink : 30% F.S.	0.853	0.087	0.812 to 0.894	0.711	0.969

No significant differences were observed.

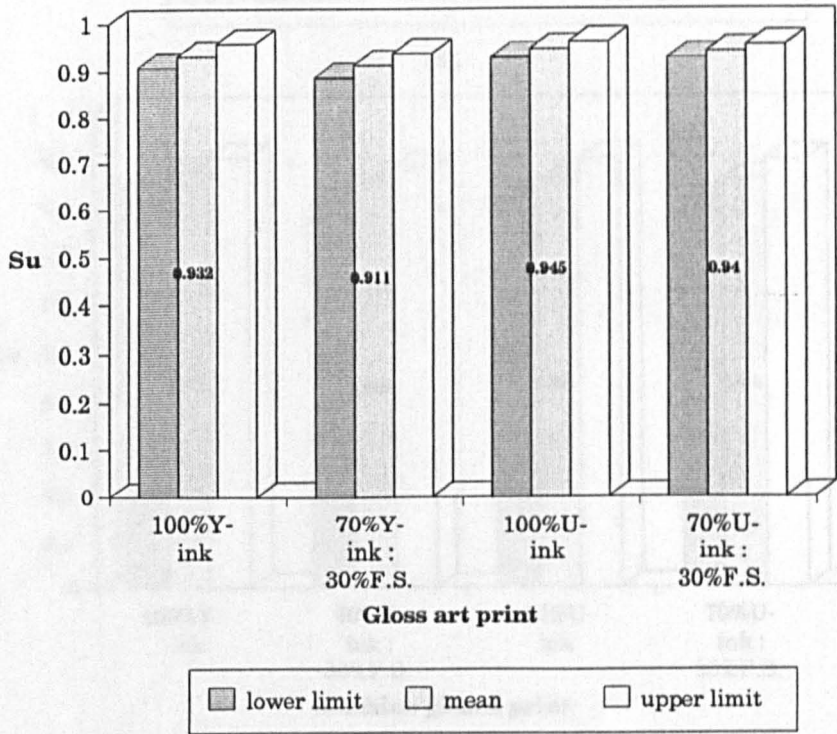
Figure 4.18 Print smoothness of gloss art (pigmented and unpigmented prints).

(a) Macrosmoothness.

(b) Microsmoothness.



(a)

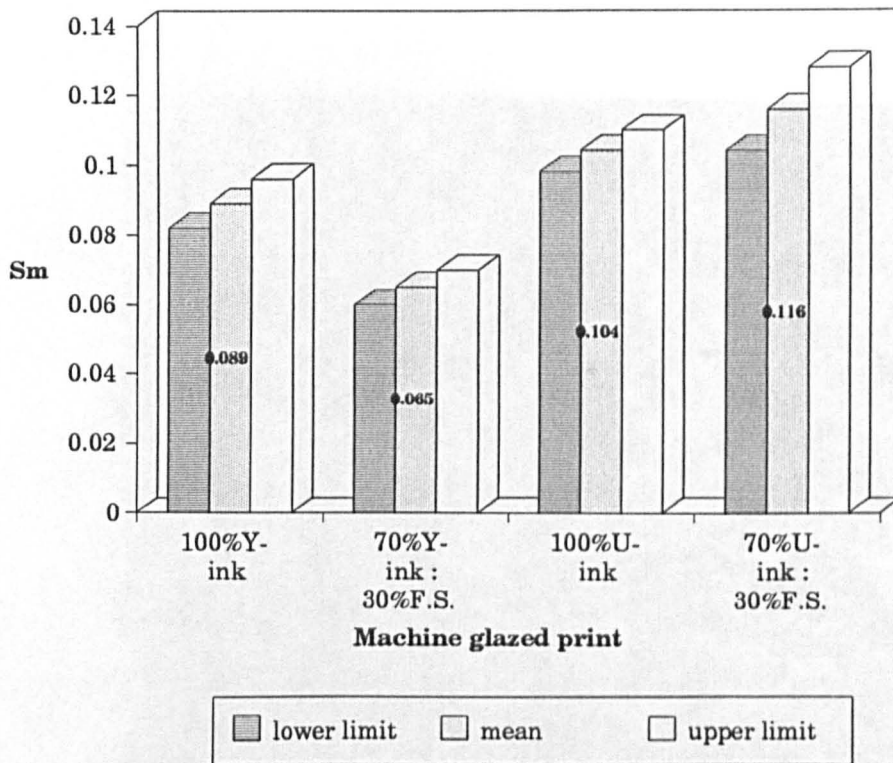


(b)

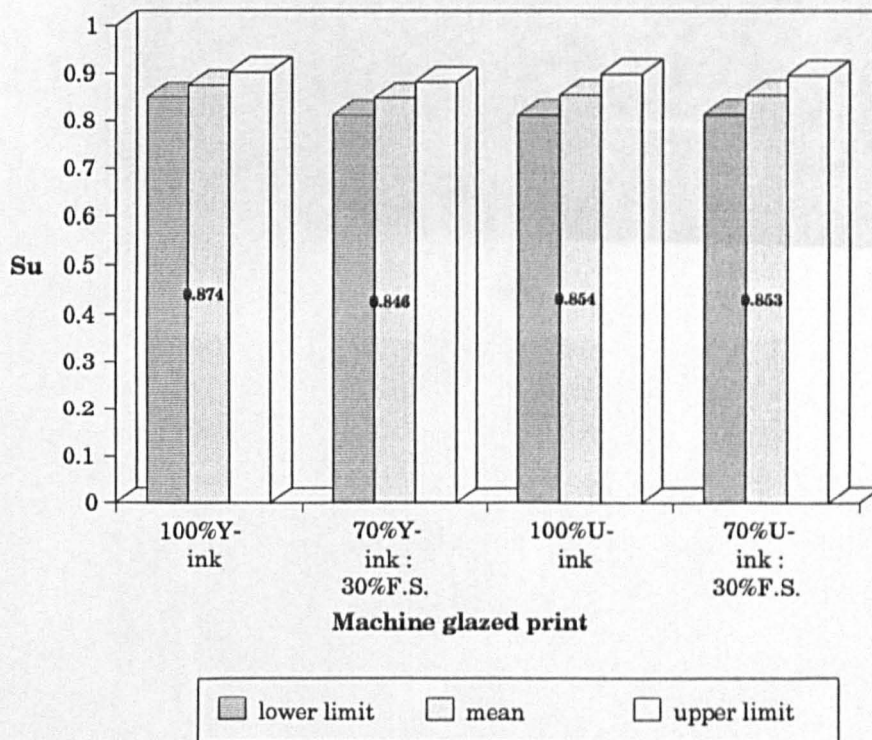
Figure 4.19 Print smoothness of machine glazed  
(pigmented and unpigmented prints).

(a) Macrosmoothness.

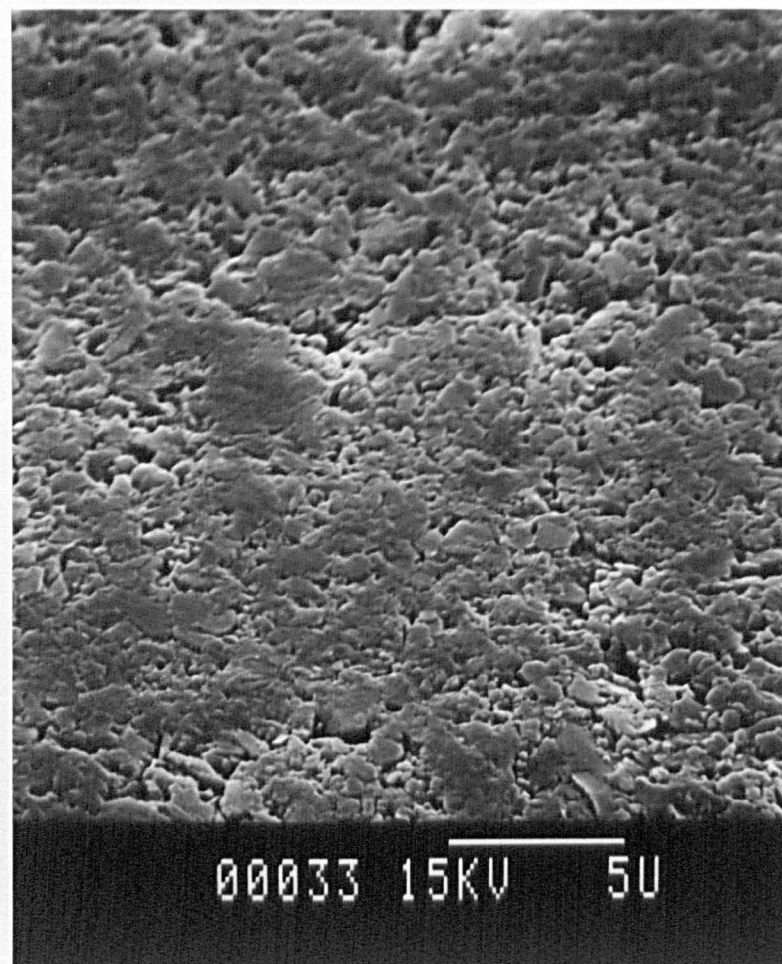
(b) Microsmoothness.



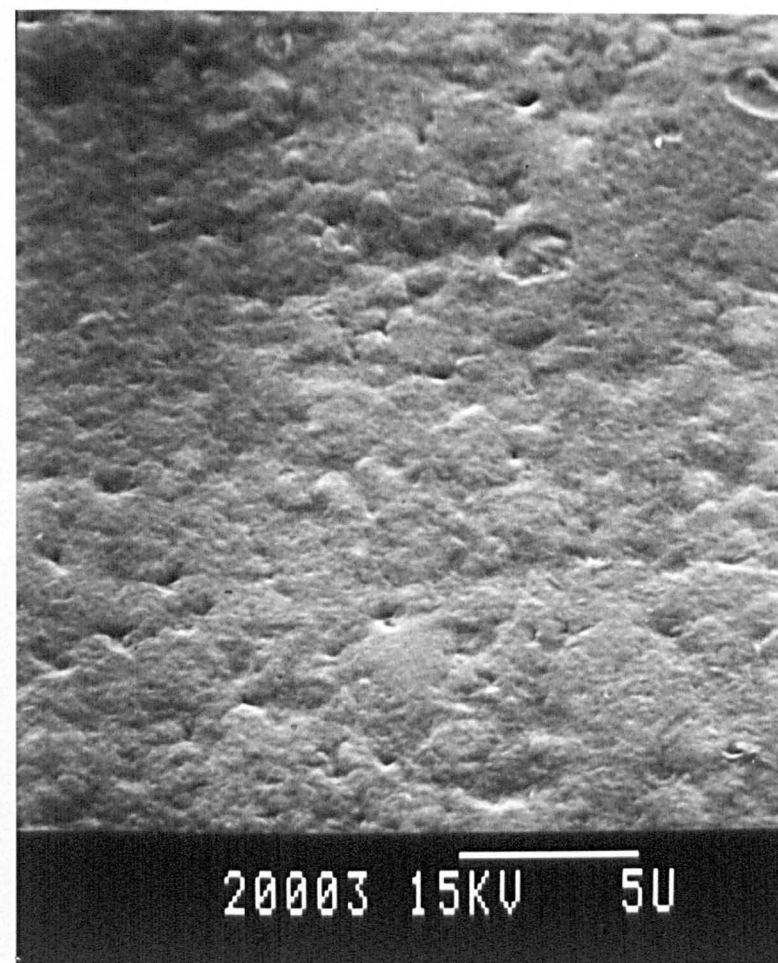
(a)



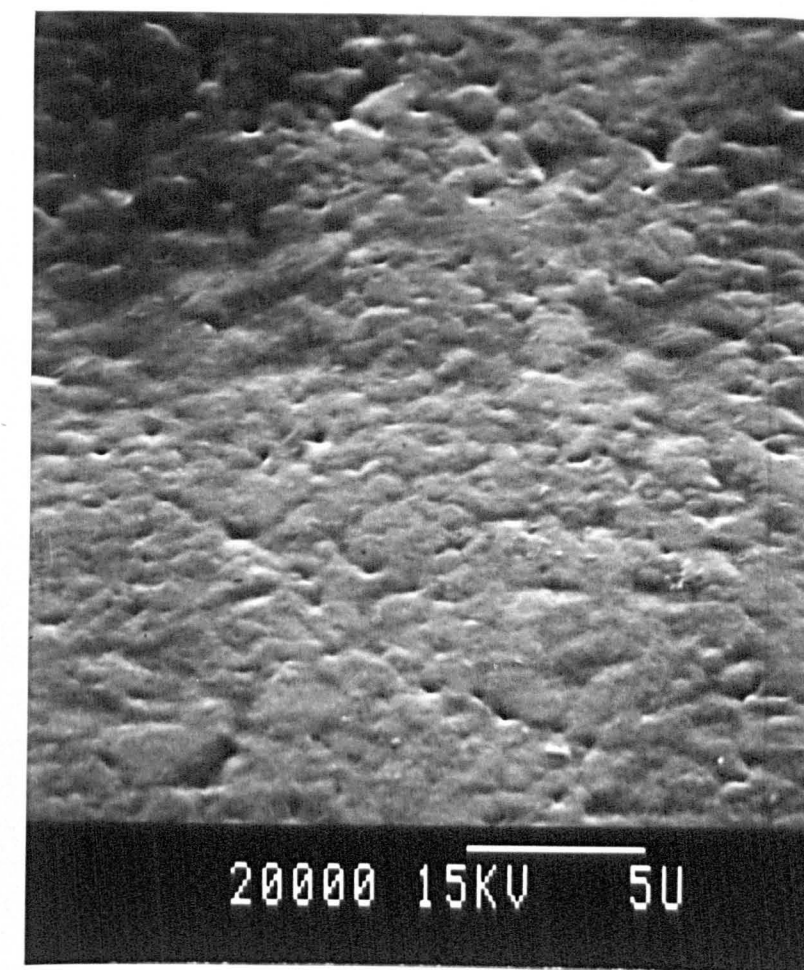
(b)



(a)



(b)



(c)

**Figure 4.20 Scanning electron micrographs x 5000.**

**(a) unprinted GA.**

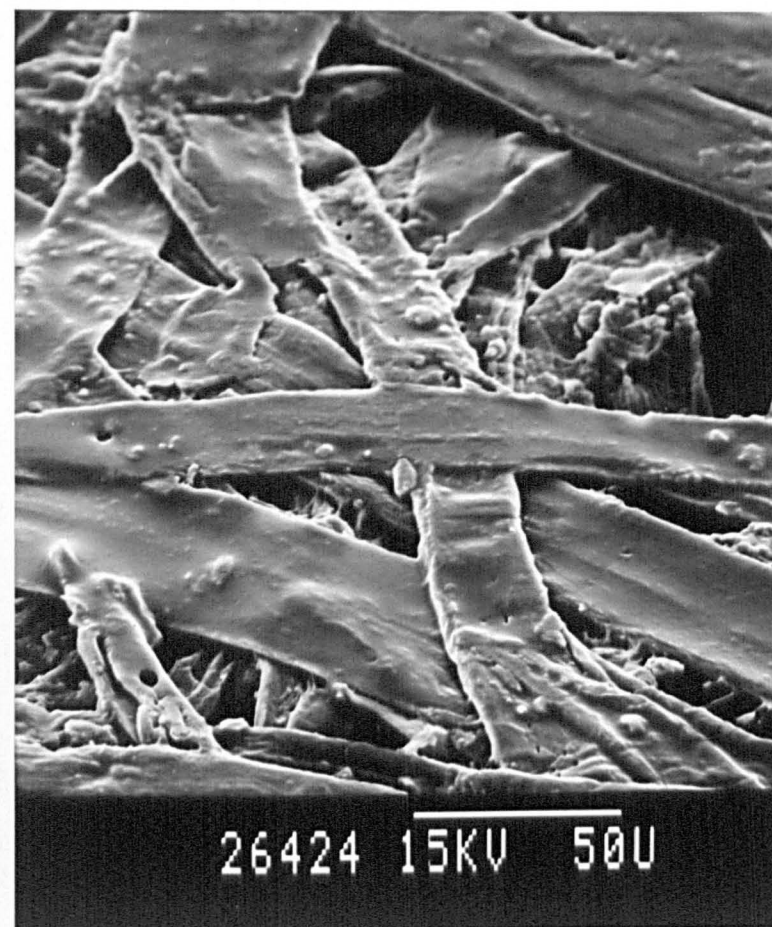
**(b) GA printed with 100%U-ink.**

**(c) GA printed with 70%U-ink:30%F.S.**

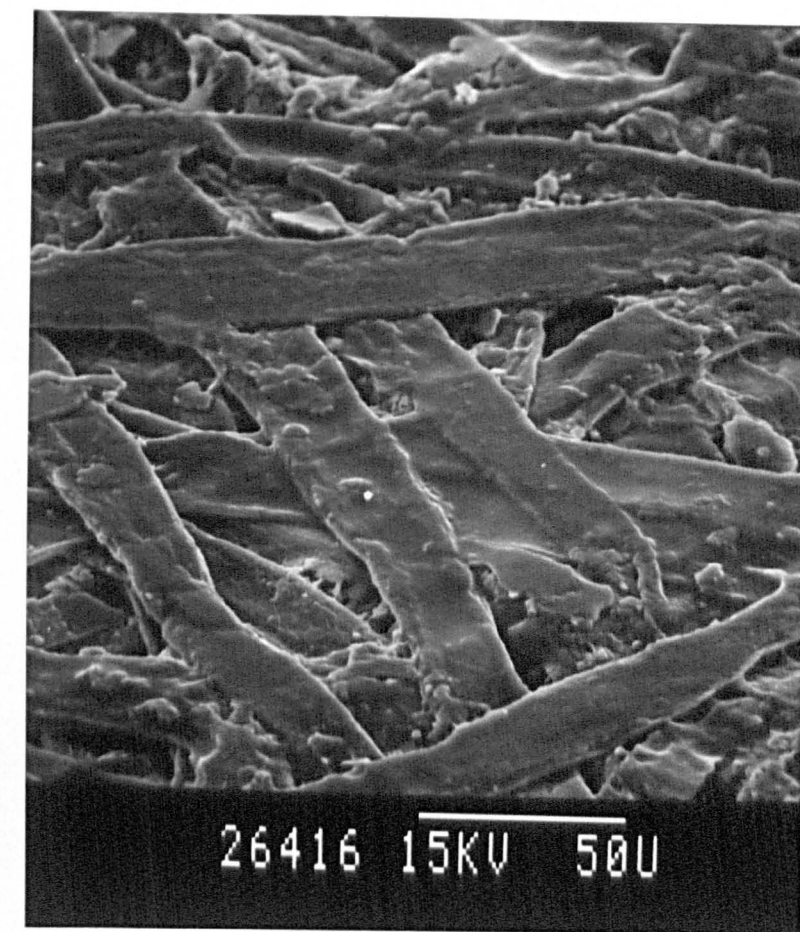




(a)



(b)



(c)

**Figure 4.21 Scanning electron micrographs x 600.**

**(a) unprinted MG.**

**(b) MG printed with 100%U-ink.**

**(c) MG printed with 70%U-ink:30%F.S.**

#### 4.3.6 Discussion - Part II(b).

The result of both gloss art and machine glazed printed with 30% water content 'unpigmented emulsion ink' indicates a smoother surface than that printed with unpigmented ink (no emulsified fountain solution) although the increase in the case of gloss art is not significant. To explain this effect, it is necessary to consider the emulsification behaviour of the unpigmented system. In Figure 4.17, besides the rapid rate of emulsification within the first minute, the results show that although the rate of emulsification decreases at first, nevertheless, subsequently the rate remains fairly constant and does not tend to zero; which means that the unpigmented ink has not yet reached its saturation for emulsified fountain solution. It is capable of taking up more fountain solution. Therefore, it is postulated that with 30% water content in this unpigmented ink, the ink is enabled to form fine emulsified fountain solution droplets of a narrow range of droplet size. These emulsified droplets enhance cavitation, as previously described, and consequently the ink transfers more evenly.

It is also found that all prints, printed with unpigmented ink and its (unpigmented) emulsion ink, have smoother surfaces than those printed with heatset yellow ink and 'yellow emulsion inks'. Thus, it is interesting to compare the emulsification behaviour between heatset yellow ink and unpigmented ink. In Figure 4.2 and Figure 4.17, it can be seen that within the first minute of emulsification, the pigmented system has emulsified approximately three times the amount as compared with the unpigmented system. However, the rate of emulsification for the pigmented system first drops dramatically and tends to zero which means that the pigmented system reaches saturation for emulsified fountain solution. This is not the case for

the unpigmented system as we have seen. It can be assumed that the system which easily reaches its saturation for emulsified water tends to have a higher probability for emulsified droplets to amalgamate. Therefore, it may be inferred that the unpigmented ink may form fine emulsified droplets of a narrower range in size; as compared to heatset yellow ink, having a wider range in size. The 'unpigmented emulsion ink' then transfers more evenly. A conclusion therefore can be drawn that the amount of emulsified fountain solution is not solely responsible for the decrease in print smoothness. The nature of the 'emulsion ink' having pigments in it is a more dominant contributor to this decrease. Pigment aggregates may occur which, as previously discussed, can lead to uneven ink film formation.

With reference to the proposed printed surface model, it can be understood that the decrease in print smoothness of both gloss art and machine glazed prints is mainly due to a change in average facet angles at the area where the pigment aggregates adhere and to a smaller extent due to the fibre and/or the coating rising.

Referring to the scanning electron micrographs in Figures 4.20-4.21, because where no pigment is included in the ink, it might be erroneous to draw a conclusion *a priori* that these ink films have the same contours as the unprinted surfaces. However careful consideration of the images suggests that these 'unpigmented' printed surfaces are uniform ink films which follow the details of the unprinted surfaces underneath.

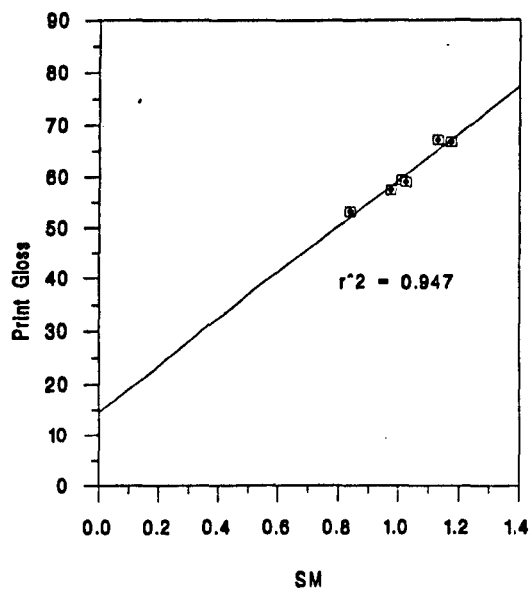
It is interesting to note that the results from this study coincide with the results from other studies [Hayashi & Amari, 1992; Bery & Loel, 1992].

Hayashi & Amari studied dynamic ink transfer and splitting of emulsified ink and found that emulsified fountain solution in the ink accelerates the formation of pigment aggregates. Bery & Loel studied eight coated papers printed on a commercial web offset press and evaluated surface roughness of both unprinted and printed surfaces using a Parker Print-surf instrument. They found in a single colour printing that emulsification of fountain solution in the ink caused a decrease in the print smoothness. The results in the present study, as observed by the optical method also indicate a decrease in print smoothness. This confirms that, where print smoothness is of importance and is to be studied, an appropriate laboratory set-up using this optical method can yield reliable results which can be correlated to practical printing. This helps to save the cost of using an expensive commercial printing press.

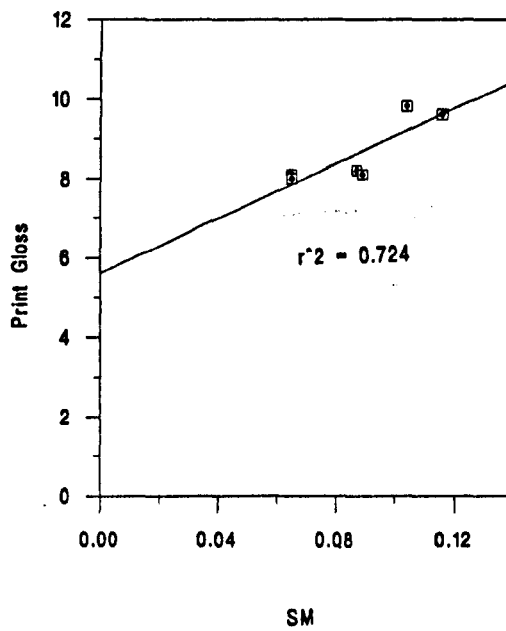
### **Relationship between print gloss and print smoothness.**

The earlier study of paper surface structure [Hansuebsai, 1989] has found that gloss correlates well with macrosmoothness. The relation between print gloss and print smoothness is also studied. Figure 4.22 shows the plot between print gloss from Tables 4.6, 4.12 and their macrosmoothness values. In the case of the gloss art prints the value for  $R^2$  is 0.95 while  $R^2$  is only 0.72 for the machine glazed prints. The results show a better correlation between print gloss and print macrosmoothness for gloss art prints than for machine glazed prints. This is due to the fact that the gloss values of machine glazed prints are very low. This indicates that the geometry of the gloss meter employed in the study is not appropriate for low gloss surfaces. It was found [Fetsko & Zettlemoyer, 1962] from prints made with one ink on two papers (uncoated and coated papers) that coated prints produced a high narrow peak





(a)



(b)

**Figure 4.22 Print gloss vs print smoothness.**

**(a) gloss art samples.**

**(b) machine glazed samples.**

at the specular angle whereas the maximum reflectance for uncoated prints was shifted towards a larger angle than the specular angle. This leads to the conclusion that the optimum geometrical condition varies for prints made with different ink-paper combinations, whereas a standard optimum geometrical condition is necessary for a reliable comparison.

The present study also attempted to evaluate print roughness by using a Parker Print-surf instrument (PPS automatic version 1.1) in order to find a relationship between PPS print roughness and the optical print smoothness. However, the width of the prints obtained on the laboratory IGT printability tester was 32 mm which is smaller than the diameter of the measuring area of 35 mm required by the PPS. Hence, the PPS print roughness test was not performed.

### **Implications for print mottle.**

When an ink fails to form a smooth uniform film after ink transfer, mottle can occur. Print mottle is defined as an uneven ink distribution layer of solid area on the printed surface and it is one of the most commonly occurring print defects in the offset lithographic process. Goodman [1992] stated that emulsification in the ink was one of the major causes of print mottle. Bassemir *et al.* [NPIRI, 1990] studied the effects of ink water pickup on several aspects of print quality including print mottle using a high speed web offset lithographic press. They determined print mottle by measuring optical print density and used the standard deviation in optical print density as an indicator of print mottle. They found that a high water pickup ink showed high print mottle at low and high water feed conditions. The present study is relevant to those studies since one type of print mottle may be

defined as pigment mottle [Carlsson & Lindberg, 1971]. Carlsson & Lindberg studied print mottle in a printing press using high-speed photography; they found that print mottle occurs in different ways distinguished as pigment mottle, displacement mottle and non-wetting mottle. It is noted that most methods of print mottle measurement are only qualitatively based on visual observation. The potential use of print smoothness as a direct indicator for print mottle is, therefore, of interest. Further analysis of print smoothness data may be useful to provide a standard indicator for print mottle.

### **Limitations in surface characterization techniques used.**

It has to be accepted that the methods employed in the present study namely print gloss and scanning electron microscopy, each has its limitations. The scanning electron microscope (SEM) has been extensively used to give high magnification visual images of a surface. Although the wide magnification range of SEM makes the method capable of recording the detailed features of both macrosmoothness and microsmoothness, the micrographs only give qualitative information which requires good knowledge and experience of the observers to interpret. Such assessments therefore depend on the observers' perception. Gloss meters, on the other hand, generally measure smoothness at only one angle of incidence and reflectance, whether macrosmoothness or microsmoothness. Moreover, as discussed, the optimum geometrical condition varies for each combination of ink and paper; comparisons between print gloss can be made only if a standard geometrical condition is defined. The optical method is found useful as it can evaluate the whole surface. However, the present instrument needs further improvements to enable automated operation. Its present manual mode is too time consuming for routine work.

## **4.4 CONCLUSIONS AND FURTHER WORK.**

### **4.4.1 Conclusions.**

An optical reflectance instrument, a type of goniophotometer was specially rebuilt and utilized to measure surface reflection for both paper and printed paper surfaces. The work confirmed that the improved instrument was adequate for studies of printed paper surfaces; where the previous instrument could cope only with paper surfaces. The reflectance data were subjected to correlation and regression analysis to yield optical smoothness parameters, namely macrosmoothness and microsmoothness. The optical smoothness results obtained were statistically analysed to determine whether the results are significantly affected by a range of experimental treatments. Assessments are made in terms of 95% confidence limits.

It should be recalled that the results from the preliminary study have shown that different papers were affected in differing ways by various 'ink vehicles'. The most significant, contrasting, changes were found with machine glazed and poladin cartridge paper. Ink vehicles, R1006 and SM2007, caused an increase in both macrosmoothness and microsmoothness of poladin cartridge. In contrast, these ink vehicles each caused a decrease in both macrosmoothness and microsmoothness of machine glazed. Consequently, it was decided to choose two contrasting types of paper, namely gloss art and machine glazed, for the study of print smoothness phenomena.

Based on the fact that most inks generally consist of pigment, ink vehicle and additives of which the major proportion is the ink vehicle, a printed surface model is proposed as an ink vehicle-air interface rather than

a pigment-air interface. A printed surface is a result of material interactions in a printing process; and this model appears to lead to a better understanding of ink and paper interactions where such interactions can modify the air interface of the ink film.

Printed surface characteristics of gloss art were found to be no less smooth than its paper surface characteristics. The macrosmoothness and microsmoothness values for both gloss art paper and printed gloss art are high, within a range of approximately 0.90-1.20. On the contrary, the printed surface characteristics of machine glazed was found to be rougher than its paper surface characteristics; although a decrease was shown in the macrosmoothness values only. The macrosmoothness decreased from a value of 0.11 down to 0.09. These two low smoothness values indicate the roughness of both the paper and the printed surfaces of machine glazed. It can be seen that the development of printed surface characteristics is affected by various phenomena in ink/paper interactions to a different extent. The first phenomenon namely ink/paper contact is seen to be less significant due to the use of a resilient blanket in the offset lithographic process. Ink penetration and ink film splitting are more important phenomena affecting these printed surfaces. The penetration of ink into paper also plays a part in the ink drying process. The contribution of the paper properties is more dominant in these phenomena as it is found that the amount of ink transfer for an uncoated paper is almost twice that for a coated paper. A conclusion drawn from this finding is that ink penetration plays a more important role in determining the printed surface characteristics of uncoated papers; whereas ink film splitting is a more dominant factor for coated papers. Separation of the ink vehicle from the pigments, a well-known filtration

phenomenon, was clearly seen in the case of gloss art paper. This indicates the significant role of porous structure within the paper and the role of pigment particle sizes within the ink.

The interactions between 'emulsion ink' and paper which occur in the offset lithographic process are not significantly different from interactions in other printing processes, where water does not play a role. Printed surface characteristics are developed through the same phenomena. However, because fountain solution is incorporated in the lithographic ink, the printed surfaces are found to behave differently, leading to rougher surfaces for both uncoated and coated paper samples. Print smoothness decreased when the amount of emulsified fountain solution is greater than a critical amount for the ink. The critical amount for the yellow ink used can be concluded to be at 15% water content in 'emulsion ink'. At both 20% and 30% water content in the 'emulsion ink', the print smoothness of machine glazed samples decreased by almost 30%; and also at 30% water content the print smoothness of gloss art decreased by almost 14%. These results demonstrated the influences of emulsification behaviour on the resultant print qualities which may arise from the various phenomena during ink transfer and ink drying processes. In addition, it was found that this emulsification behaviour was connected with the pigments. The fountain solution when emulsified in the ink is believed to spread a thin film around the pigment surfaces which eventually join up together to form pigment aggregates. Such aggregates protrude from the ink film when the ink film was dried and pigment aggregation is found to be a more important contributor to the decrease in print smoothness than the amount of the emulsified fountain solution in the ink. These findings are of significance to the offset litho ink makers who are

ink. These findings are of significance to the offset litho ink makers who are required to produce high pigmentation inks, owing to the nature of the offset lithographic process. These findings are also beneficial to the printer who needs to rapidly adjust the printing conditions to obtain best resultant prints with different combinations of ink and paper.

In conclusion, this optical method makes it possible to evaluate quality of print in terms of print smoothness by employing a suitable laboratory test; prior to using a commercial printing press which would be costly and time consuming. In addition, this method also provides a means to obtain more direct data on the factors that affect the quality of printed surfaces.

#### **4.4.2 Further work.**

An ink will dramatically increase print smoothness and print gloss of uncoated papers whereas it can impair print gloss by disturbing the inherent coated paper gloss. It would be interesting for further work to investigate the influences of varying printing conditions such as ink film thickness, printing speed on print smoothness for different types of paper and using different pigment particle sizes, especially for offset lithography where the effect of paper swelling on pore dimension may be significant. The results may lead to a better understanding of various phenomena involved in ink and paper interactions, for example, the contact and the splitting of an ink film with increasing ink film thickness and with decreasing ink film thickness.

In this study, the reflectance measurement and the data collection and analysis necessary to obtain the optical smoothness, were carried out manually. This is a tedious method particularly when a large number of samples are examined. The method needs to be improved by means of

motorized control and computerized data collection and analysis. This would help to cut down the testing time so that the results for surfaces under investigation can be obtained quicker and eventually some problems may be solved better.

In addition, the ink chosen for study was a yellow ink. This is consistent with the use of a 632.8 nm HeNe laser as the light source of the reflectance instrument. To further establish the validity of this method to other colours of inks, the use of a dual colour laser namely a red/infrared laser is proposed. The visible red output would be used to set up the instrument. A filter could then be used to block the 632.8 nm line leaving the invisible line correctly aligned through the instrument. The longer infrared wavelength may provide interesting information on microsmoothness because the behaviour of the reflectance of the microfeatures would be closer to that of Fresnel reflectance.



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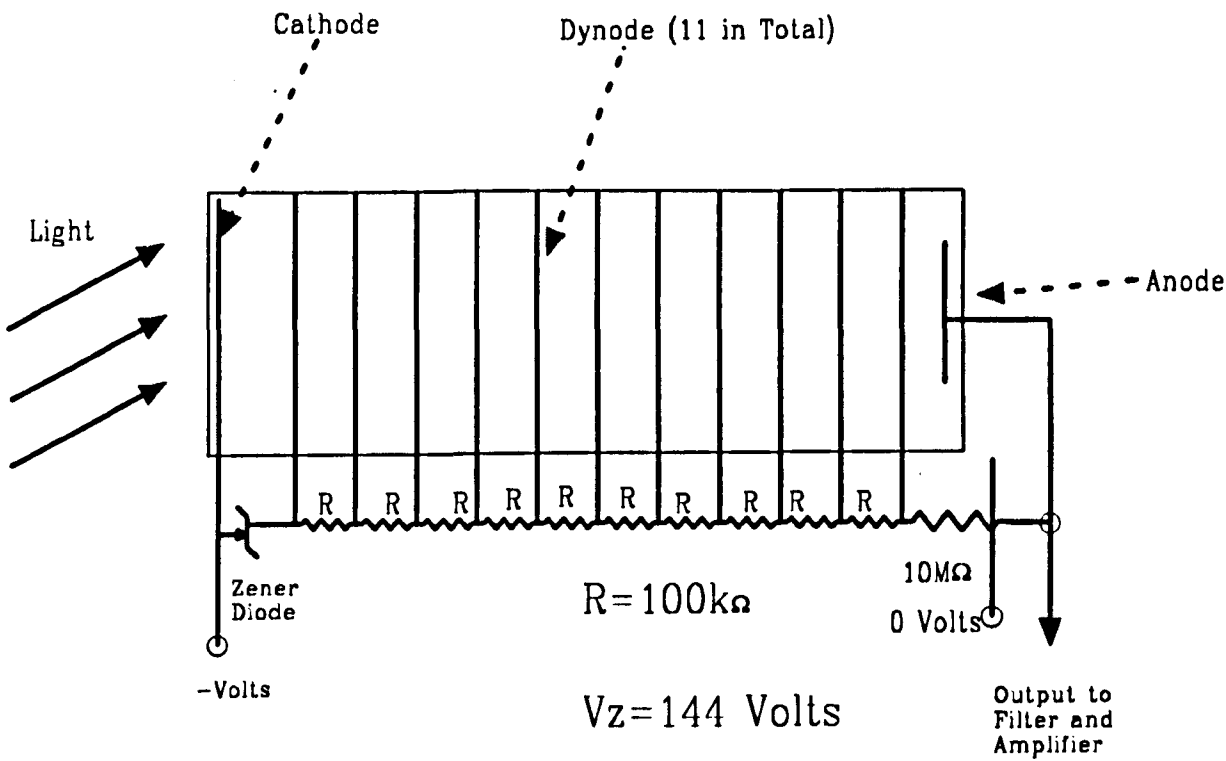
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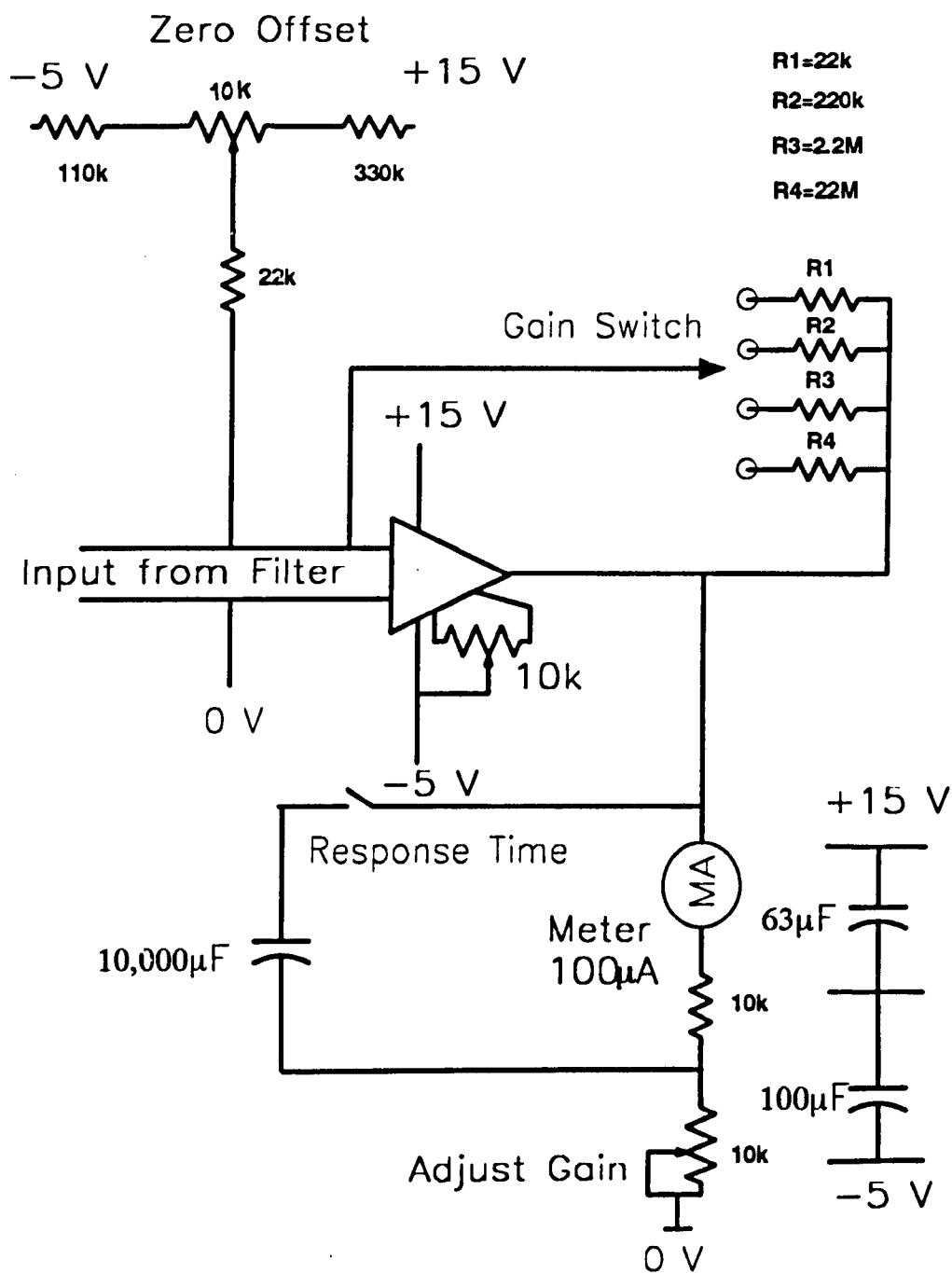
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Appendix A: Figure A.1



Circuit of Photomultiplier

## Appendix A: Figure A.2



**Circuit of Amplifier /Meter**



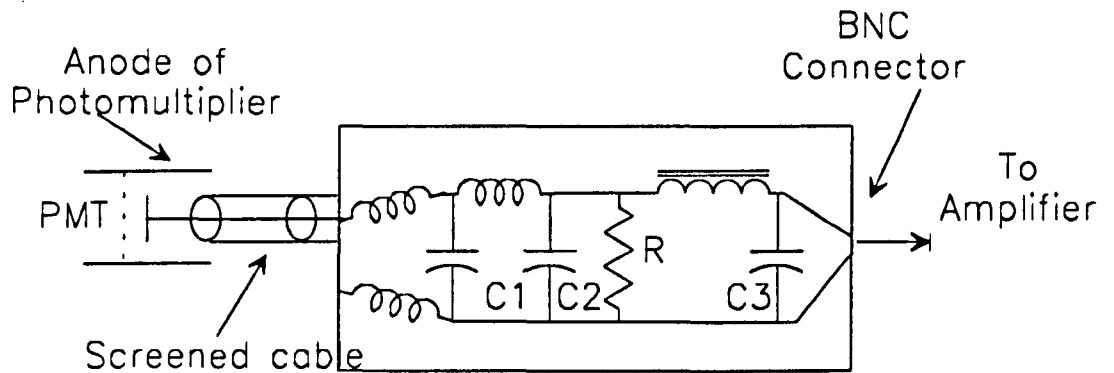
## Appendix A: Figure A.3

$$C1 = 0.01\mu\text{F}$$

$$C2 = 0.47\mu\text{F}$$

$$C3 = 10.0\mu\text{F}$$

$$R = 1\text{ M}\Omega$$



**Circuit of a Low-Pass Filter**

## Appendix B.

Table B.1 Fresnel reflectance for refractive indices (n) from 1.5-1.9 according to equation 21 in section 2.6.3.

Angle	n=1.5	n=1.55	n=1.6	n=1.7	n=1.8	n=1.9
15	4.38	5.08	5.80	7.29	8.81	10.35
20	4.71	5.45	6.20	7.76	9.35	10.95
25	5.16	5.95	6.76	8.41	10.08	11.76
30	5.78	6.64	7.51	9.28	11.06	12.84
35	6.61	7.55	8.51	10.42	12.34	14.23
40	7.72	8.77	9.82	11.92	13.99	16.01
45	9.20	10.38	11.55	13.86	16.11	18.28
50	11.21	12.54	13.85	16.40	18.84	21.18
55	13.93	15.43	16.90	19.71	22.36	24.86
60	17.66	19.35	20.99	24.06	26.91	29.55
65	22.81	24.69	26.48	29.79	32.79	35.53
70	29.96	31.99	33.89	37.34	40.40	43.14
75	39.94	42.01	43.91	47.30	50.24	52.83
80	53.86	55.74	57.44	60.42	62.93	65.11

Table B.2 Slope values of fresnel reflectance for refractive indices from 1.5-1.9 and the averaged slope values used as the standard.

<b>Region</b>	<b>n=1.5</b>	<b>n=1.6</b>	<b>n=1.7</b>	<b>n=1.8</b>	<b>n=1.9</b>	<b>standard</b>
15-20	0.07	0.08	0.09	0.11	0.12	0.09
20-25	0.09	0.11	0.13	0.15	0.16	0.13
25-30	0.12	0.15	0.17	0.20	0.22	0.17
30-35	0.17	0.20	0.23	0.26	0.28	0.23
35-40	0.22	0.26	0.30	0.33	0.36	0.29
40-45	0.30	0.35	0.39	0.42	0.45	0.38
45-50	0.40	0.46	0.51	0.55	0.58	0.50
50-55	0.54	0.61	0.66	0.70	0.74	0.65
55-60	0.75	0.82	0.87	0.91	0.94	0.86
60-65	1.03	1.10	1.15	1.18	1.20	1.13
65-70	1.43	1.48	1.51	1.52	1.52	1.49
70-75	2.00	2.00	1.99	1.97	1.94	1.98
75-80	2.79	2.71	2.62	2.54	2.46	2.62

## Appendix C.

Table C.1 Macrosmoothness (Sm) results of newsprint samples before and after being treated with ink 'vehicles' (See Table 3.1a).

Sample		NP / Sm			
No.	Untreated	R1066	R1976	R1835	SM2007
1	0.008	0.022	0.015	0.022	0.008
2	0.015	0.023	0.024	0.023	0.008
3	0.009	0.014	0.023	0.022	0.014
4	0.010	0.016	0.015	0.019	0.009
5	0.010	0.015	0.020	0.022	0.016
6	0.013	0.018	0.018	0.018	0.020
7	0.010	0.021	0.016	0.015	0.011
8	0.012	0.016	0.013	0.020	0.017
9	0.013	0.017	0.018	0.024	0.011
<b>X</b>	0.011	0.018	0.021	0.018	0.013
<b>S.D.</b>	0.002	0.003	0.003	0.004	0.004

Table C.2 Microsmoothness (Su) results of newsprint samples before and after being treated with ink 'vehicles' (See Table 3.1b).

Sample		NP / Su			
No.	Untreated	R1066	R1976	R1835	SM2007
1	0.619	0.770	0.768	0.688	0.613
2	0.757	0.789	0.692	0.796	0.936
3	0.753	0.892	0.725	0.750	0.625
4	0.832	0.750	0.819	0.742	0.773
5	0.695	0.876	0.711	0.767	0.939
6	0.773	0.808	0.685	0.715	0.741
7	0.642	0.898	0.799	0.738	0.866
8	0.698	0.778	0.738	0.809	0.779
9	0.782	0.803	0.649	0.690	0.917
$\bar{X}$	0.728	0.818	0.744	0.732	0.799
S.D.	0.069	0.056	0.043	0.055	0.126

Table C.3 Macrosmoothness (Sm) results of machine finish samples before and after being treated with ink 'vehicles' (See Table 3.2a).

Sample No.	MF / Sm			
	Untreated	R1066	R1835	SM2007
1	0.007	0.009	0.007	0.021
2	0.006	0.012	0.009	0.027
3	0.010	0.018	0.006	0.022
4	0.012	0.019	0.017	0.022
5	0.011	0.021	0.006	0.017
6	0.008	0.011	0.006	0.020
7	0.009	0.010	0.006	0.017
8	0.008	0.020	0.005	0.017
9	0.009	0.014	0.011	0.021
$\bar{X}$	0.009	0.015	0.008	0.020
S.D.	0.002	0.005	0.004	0.003

Table C.4 Microsmoothness (Su) results of machine finish samples before and after being treated with ink 'vehicles' (See Table 3.2b).

Sample	MF / Su			
	Untreated	R1066	R1835	SM2007
1	0.199	0.635	0.388	0.613
2	0.168	0.812	0.343	0.776
3	0.162	0.768	0.487	0.730
4	0.231	0.729	0.454	0.758
5	0.215	0.750	0.456	0.694
6	0.234	0.805	0.369	0.604
7	0.265	0.736	0.397	0.730
8	0.235	0.787	0.534	0.802
9	0.186	0.629	0.197	0.785
$\bar{X}$	0.211	0.739	0.403	0.721
S.D.	0.034	0.067	0.098	0.072

Table C.5 Macrosmoothness (Sm) results of machine glazed samples before and after being treated with ink 'vehicles' (See Table 3.3a).

Sample No.	MG / Sm		
	Untreated	R1066	SM2007
1	0.108	0.025	0.062
2	0.067	0.035	0.099
3	0.072	0.053	0.042
4	0.069	0.048	0.037
5	0.077	0.052	0.043
6	0.069	0.035	0.047
7	0.082	0.041	0.064
8	0.070	0.040	0.037
9	0.068	0.048	0.040
$\bar{X}$	0.076	0.042	0.052
S.D.	0.013	0.009	0.020



Table C.6 Microsmoothness (Su) results of machine glazed samples before and after being treated with ink 'vehicles' (See Table 3.3b).

Sample No.	MG / Su		
	Untreated	R1066	SM2007
1	0.766	0.970	0.842
2	0.899	0.920	0.727
3	0.822	0.862	0.816
4	0.942	0.931	0.799
5	0.941	0.764	0.743
6	0.903	0.945	0.773
7	0.915	0.776	0.880
8	0.949	0.863	0.859
9	0.934	0.832	0.781
$\bar{X}$	0.897	0.874	0.802
S.D.	0.062	0.074	0.052

Table C.7 Macrosmoothness (Sm) results of poladin cartridge samples before and after being treated with ink 'vehicles' (See Table 3.4a).

Sample No.	PC / Sm		
	Untreated	R1066	SM2007
1	0.007	0.019	0.049
2	0.009	0.009	0.045
3	0.009	0.019	0.061
4	0.013	0.020	0.065
5	0.008	0.022	0.047
6	0.009	0.017	0.052
7	0.012	0.018	0.031
8	0.009	0.019	0.053
9	0.012	0.023	0.040
<b>X̄</b>	0.010	0.018	0.049
<b>S.D.</b>	0.002	0.004	0.010

Table C.8 Microsmoothness (Su) results of poladin cartridge samples before and after being treated with ink 'vehicles' (See Table 3.4b).

Sample No.	PC / Su		
	Untreated	R1066	SM2007
1	0.151	0.777	0.930
2	0.180	0.606	0.859
3	0.137	0.805	0.867
4	0.164	0.932	0.665
5	0.135	0.648	0.769
6	0.203	0.884	0.772
7	0.212	0.753	0.897
8	0.157	0.845	0.858
9	0.144	0.884	0.789
$\bar{X}$	0.165	0.793	0.823
S.D.	0.028	0.110	0.081

## Appendix D.

Table D.1

(a) Summary of analysis of variance for macrosmoothness (Sm) of newsprint samples.

Source of Variation	D.F.	Sum of Squares	Mean Squares	F Ratio
Between Groups	4	0.0006	0.0001	12.7739
Within Groups	40	0.0004	0.0000	
Total	44	0.0010		

The F ratio was compared to the value of the F distribution with 4 and 40 degrees of freedom, which is approximately 2.61. This indicates that the macrosmoothness values between these newsprint samples are significantly different (see Table 3.1a).

(b) Multiple comparison test - Duncan procedure for Sm of newsprint samples.

	NP + R1066	NP + R1976	NP + R1835	NP + SM2007
Untreated NP	*	*	*	
NP + R1066				
NP + R1976				
NP + R1835				
NP + SM2007	*	*	*	

\* Denotes pair of samples with a significant difference at the 0.05 level (see Table 3.1a).

Table D.2

(a) Summary of analysis of variance for microsmoothness (Su) of newsprint samples.

Source of Variation	D.F.	Sum of Squares	Mean Squares	F Ratio
Between Groups	4	0.0621	0.0155	2.7160
Within Groups	40	0.2285	0.0057	
Total	44	0.2906		

The F ratio was compared to the value of the F distribution with 4 and 40 degrees of freedom, which is approximately 2.61. This indicates that the microsmoothness values between these newsprint samples are significantly different (see Table 3.1b).

(b) Multiple comparison test - Duncan procedure for Su of newsprint samples.

	NP + R1066	NP + R1976	NP + R1835	NP + SM2007
Untreated NP	*			
NP + R1066				
NP + R1976				
NP + R1835	*			
NP + SM2007				

. \* Denotes pair of samples with a significant difference at the 0.05 level (see Table 3.1b).

Table D.3

(a) Summary of analysis of variance for macrosmoothness (Sm) of machine finish samples.

Source of Variation	D.F.	Sum of Squares	Mean Squares	F Ratio
Between Groups	3	0.0009	0.0003	23.7713
Within Groups	32	0.0004	0.0000	
Total	35	0.0013		

The F ratio was compared to the value of the F distribution with 3 and 32 degrees of freedom, which is approximately 2.90. This indicates that the macrosmoothness values between these machine finish samples are significantly different (see Table 3.2a).

(b) Multiple comparison test - Duncan procedure for Sm of machine finish samples.

	MF + R1066	MF + R1835	MF + SM2007
Untreated MF	*		*
MF + R1066			*
MF + R1835	*		*
MF + SM2007			

\* Denotes pair of samples with a significant difference at the 0.05 level (see Table 3.2a).

Table D.4

(a) Summary of analysis of variance for microsmoothness (Su) of machine finish samples.

Source of Variation	D.F.	Sum of Squares	Mean Squares	F Ratio
Between Groups	3	1.7818	0.5939	116.0313
Within Groups	32	0.1638	0.0051	
Total	35	1.9457		

The F ratio was compared to the value of the F distribution with 3 and 32 degrees of freedom, which is approximately 2.90. This indicates that the microsmoothness values between these machine finish samples are significantly different (see Table 3.2b).

(b) Multiple comparison test - Duncan procedure for Su of machine finish samples.

	MF + R1066	MF + R1835	MF + SM2007
Untreated MF	*	*	*
MF + R1066			
MF + R1835	*		*
MF + SM2007			

\* Denotes pair of samples with a significant difference at the 0.05 level (See Table 3.2b).

**Table D.5**

(a) Summary of analysis of variance for macrosmoothness (Sm) of machine glazed samples.

<b>Source of Variation</b>	<b>D.F.</b>	<b>Sum of Squares</b>	<b>Mean Squares</b>	<b>F Ratio</b>
Between Groups	2	0.0054	0.0027	12.2927
Within Groups	24	0.0053	0.0002	
Total	26	0.0107		

The F ratio was compared to the value of the F distribution with 2 and 24 degrees of freedom, which is approximately 3.40. This indicates that the macrosmoothness values between these machine glazed samples are significantly different (see Table 3.3a).

(b) Multiple comparison test - Duncan procedure for Sm of machine glazed samples.

	<b>MG + R1066</b>	<b>MG + SM2007</b>
Untreated MG	*	*
MG + R1066		
MG + SM2007		

\* Denotes pair of samples with a significant difference at the 0.05 level (see Table 3.3a).



Table D.6

(a) Summary of analysis of variance for microsmoothness (Su) of machine glazed samples.

Source of Variation	D.F.	Sum of Squares	Mean Squares	F Ratio
Between Groups	2	0.0437	0.0219	5.4636
Within Groups	24	0.0961	0.0040	
Total	26	0.1398		

The F ratio was compared to the value of the F distribution with 2 and 24 degrees of freedom, which is approximately 3.40. This indicates that the microsmoothness values between these machine glazed samples are significantly different (see Table 3.3b).

(b) Multiple comparison test - Duncan procedure for Su of machine glazed samples.

	MG + R1066	MG + SM2007
Untreated MG		*
MG + R1066		*
MG + SM2007		

\* Denotes pair of samples with a significant difference at the 0.05 level (see Table 3.3b).

**Table D.7**

(a) Summary of analysis of variance for macrosmoothness (Sm) of poladin cartridge samples.

<b>Source of Variation</b>	<b>D.F.</b>	<b>Sum of Squares</b>	<b>Mean Squares</b>	<b>F Ratio</b>
Between Groups	2	0.0077	0.0039	91.7768
Within Groups	24	0.0010	0.0000	
Total	26	0.0087		

The F ratio was compared to the value of the F distribution with 2 and 24 degrees of freedom, is approximately 3.40. This indicates that the macrosmoothness values between these poladin cartridge samples are significantly different (see Table 3.4a).

(b) Multiple comparison test - Duncan procedure for Sm of poladin cartridge samples.

	<b>PC + R1066</b>	<b>PC + SM2007</b>
Untreated PC	*	*
PC + R1066		*
PC + SM2007		

\* Denotes pair of samples with a significant difference at the 0.05 level (see Table 3.4a).

Table D.8

(a) Summary of analysis of variance for microsmoothness (Su) of poladin cartridge samples.

Source of Variation	D.F.	Sum of Squares	Mean Squares	F Ratio
Between Groups	2	2.4848	1.2424	191.3075
Within Groups	24	0.1559	0.0065	
Total	26	2.6407		

The F ratio was compared to the value of the F distribution with 2 and 24 degrees of freedom, which is approximately 3.40. This indicates that the microsmoothness values between these poladin cartridge samples are significantly different (see Table 3.4b).

(b) Multiple comparison test - Duncan procedure for Su of poladin cartridge samples.

	PC + R1066	PC + SM2007
Untreated PC	*	*
PC + R1066		
PC + SM2007		

\* Denotes pair of samples with a significant difference at the 0.05 level (see Table 3.4b).

## Appendix E.

Table E.1 Macrosmoothness results of unprinted gloss art and gloss art printed with 'yellow inks' (see Tables 4.7, 4.8 and 4.13).

Sample No.	GA / Sm				
	Unprinted	100%Y- ink	90%Y- ink	80%Y- ink	70%Y- ink
			+ 10% F.S.	+ 20% F.S.	+ 30% F.S.
1	1.129	0.931	0.958	1.015	0.849
2	1.077	1.005	0.959	1.037	0.880
3	0.915	0.995	1.160	1.058	0.860
4	0.873	0.855	0.885	1.058	0.943
5	0.972	0.958	0.952	1.020	0.932
6	0.910	1.088	0.933	1.070	0.745
7	0.969	0.892	0.933	0.853	0.834
8	0.928	0.942	0.900	1.043	0.816
9	0.966	1.004	1.200	0.923	0.713
10	0.922	0.892	1.103	1.019	0.668
11	1.052	0.876	1.090	0.891	0.833
12	1.109	1.133	1.160	1.079	0.937
13	0.901	0.912	1.070	1.126	0.817
14	0.952	0.904	1.088	0.929	0.928
15	0.988	0.940	0.890	1.055	0.878
16	0.873	1.079	1.210	0.985	0.872
17	0.993	1.041	0.988	0.926	0.868
18	0.979	1.011	1.071	1.225	0.875
19	0.953	0.908	0.968	0.877	0.785
20	0.901	1.122	0.939	1.001	0.730
$\bar{X}$	0.968	0.974	1.023	1.010	0.838
S.D.	0.074	0.084	0.107	0.090	0.077

Table E.2 Microsmoothness results of unprinted gloss art and gloss art printed with 'Yellow inks' (see Tables 4.7, 4.8 and 4.13).

Sample		GA / Su			
No.	Unprinted	100%Y- ink	90%Y- ink	80%Y- ink	70%Y- ink
			+ 10% F.S.	+ 20% F.S.	+ 30% F.S.
1	0.956	0.915	0.933	0.862	0.937
2	0.965	0.990	0.858	0.961	0.876
3	0.874	0.965	0.900	0.962	0.937
4	0.982	0.965	0.991	0.975	0.912
5	0.964	0.918	0.932	0.934	0.913
6	0.991	0.983	0.982	0.969	0.923
7	0.955	0.785	0.940	0.912	0.969
8	0.910	0.995	0.896	0.945	0.933
9	0.947	0.928	0.859	0.929	0.774
10	0.970	0.914	0.949	0.979	0.878
11	0.978	0.922	0.893	0.884	0.996
12	0.981	0.937	0.974	0.901	0.841
13	0.939	0.975	0.956	0.954	0.824
14	0.965	0.856	0.959	0.983	0.959
15	0.960	0.867	0.918	0.978	0.920
16	0.880	0.937	0.950	0.882	0.954
17	0.984	0.994	0.862	0.855	0.885
18	0.961	0.955	0.975	0.968	0.883
19	0.983	0.941	0.981	0.890	0.926
20	0.872	0.904	0.906	0.906	0.970
$\bar{X}$	0.951	0.932	0.931	0.932	0.911
S.D.	0.037	0.052	0.043	0.042	0.054

Table E.3 Macrosmoothness results of unprinted machine glazed and machine glazed printed with 'Yellow inks' (see Tables 4.7, 4.9 and 4.14).

Sample		MG / Sm			
No.	Unprinted	100%Y- ink	90%Y- ink	80%Y- ink	70%Y- ink
			+ 10% F.S.	+ 20% F.S.	+ 30% F.S.
1	0.113	0.090	0.079	0.069	0.076
2	0.084	0.078	0.063	0.074	0.079
3	0.123	0.083	0.091	0.066	0.064
4	0.117	0.083	0.091	0.063	0.058
5	0.112	0.116	0.078	0.063	0.060
6	0.085	0.101	0.074	0.067	0.077
7	0.095	0.084	0.119	0.066	0.071
8	0.145	0.074	0.071	0.054	0.064
9	0.117	0.073	0.063	0.037	0.045
10	0.079	0.093	0.106	0.066	0.044
11	0.125	0.071	0.073	0.071	0.053
12	0.089	0.086	0.120	0.068	0.070
13	0.093	0.126	0.061	0.047	0.062
14	0.154	0.076	0.101	0.088	0.071
15	0.101	0.102	0.067	0.073	0.087
16	0.133	0.078	0.073	0.081	0.071
17	0.108	0.078	0.099	0.072	0.066
18	0.117	0.091	0.121	0.064	0.056
19	0.123	0.087	0.090	0.052	0.064
20	0.100	0.106	0.097	0.057	0.062
$\bar{X}$	0.111	0.089	0.087	0.065	0.065
S.D.	0.020	0.015	0.020	0.012	0.011

Table E.4 Microsmoothness results of unprinted machine glazed and machine glazed printed with 'Yellow inks' (see Tables 4.7, 4.9 and 4.14).

Sample		MG / Su			
No.	Unprinted	100%Y- ink	90%Y- ink	80%Y- ink	70%Y- ink
			+ 10% F.S.	+ 20% F.S.	+ 30% F.S.
1	0.854	0.903	0.940	0.998	0.874
2	0.775	0.830	0.739	0.997	0.902
3	0.940	0.929	0.918	0.832	0.900
4	0.879	0.798	0.778	0.703	0.968
5	0.843	0.958	0.937	0.844	0.843
6	0.951	0.956	0.775	0.804	0.823
7	0.713	0.907	0.892	0.931	0.802
8	0.677	0.750	0.737	0.757	0.956
9	0.890	0.875	0.914	0.755	0.696
10	0.928	0.837	0.957	0.914	0.825
11	0.843	0.896	0.847	0.899	0.900
12	0.832	0.907	0.973	0.986	0.820
13	0.897	0.906	0.907	0.852	0.779
14	0.808	0.872	0.869	0.744	0.765
15	0.857	0.884	0.667	0.712	0.713
16	0.849	0.818	0.705	0.852	0.935
17	0.784	0.794	0.952	0.775	0.817
18	0.712	0.838	0.717	0.791	0.795
19	0.933	0.917	0.906	0.950	0.874
20	0.774	0.914	0.971	0.881	0.927
$\bar{X}$	0.837	0.874	0.855	0.849	0.846
S.D.	0.079	0.056	0.101	0.094	0.076

Table E.5 Macrossmoothness results of gloss art printed with 'unpigmented inks' (see Table 4.13).

Sample No.	GA / Sm	
	100%U-ink	70% U-ink + 30% F.S.
1	1.048	1.347
2	0.975	1.207
3	1.257	1.180
4	1.179	1.267
5	1.009	1.141
6	1.240	1.203
7	1.235	1.239
8	1.241	1.004
9	1.039	1.054
10	0.914	1.282
11	1.067	1.125
12	1.238	1.215
13	1.146	1.377
14	1.005	0.946
15	0.963	0.967
16	1.294	1.127
17	1.126	1.019
18	1.168	1.263
19	1.363	1.192
20	1.095	1.291
$\bar{X}$	1.130	1.172
S.D.	0.125	0.123



Table E.6 Microsmoothness results of gloss art printed with 'unpigmented inks' (see Table 4.13).

Sample No.	GA / Su	
	100%U-ink	70% U-ink + 30% F.S.
1	0.918	0.975
2	0.902	0.902
3	0.959	0.893
4	0.989	0.943
5	0.941	0.937
6	0.901	0.927
7	0.915	0.964
8	0.952	0.994
9	0.987	0.964
10	0.979	0.908
11	0.942	0.942
12	0.933	0.967
13	0.951	0.992
14	0.861	0.921
15	0.946	0.922
16	0.930	0.924
17	0.990	0.945
18	0.994	0.901
19	0.975	0.928
20	0.930	0.941
$\bar{X}$	0.945	0.940
S.D.	0.035	0.029

Table E.7 Macrosmoothness results of machine glazed printed with 'unpigmented inks' (see Table 4.14).

Sample No.	MG / Sm	
	100% U-ink	70% U-ink + 30% F.S.
1	0.089	0.134
2	0.103	0.102
3	0.101	0.138
4	0.116	0.097
5	0.103	0.105
6	0.096	0.083
7	0.117	0.101
8	0.103	0.169
9	0.122	0.096
10	0.093	0.114
11	0.087	0.106
12	0.114	0.00
13	0.100	0.086
14	0.116	0.150
15	0.086	0.164
16	0.111	0.087
17	0.104	0.128
18	0.086	0.122
19	0.101	0.092
20	0.132	0.147
$\bar{X}$	0.104	0.116
S.D.	0.013	0.026

Table E.8 Microsmoothness results of machine glazed printed with 'unpigmented inks' (see Table 4.14).

Sample No.	MG / Su	
	100% U-ink	70% U-ink + 30% F.S.
1	0.846	0.799
2	0.956	0.720
3	0.706	0.967
4	0.738	0.931
5	0.914	0.854
6	0.821	0.767
7	0.869	0.849
8	0.987	0.964
9	0.886	0.871
10	0.780	0.750
11	0.975	0.894
12	0.705	0.922
13	0.891	0.711
14	0.874	0.930
15	0.923	0.969
16	0.702	0.722
17	0.780	0.777
18	0.957	0.853
19	0.860	0.910
20	0.909	0.898
$\bar{X}$	0.854	0.853
S.D.	0.092	0.087

## Appendix F.

Table F.1 T-test summary: unprinted and printed with heatset yellow ink for both gloss art and machine glazed samples.

(a) Macrosmoothness (Sm).

Sample	Sm		
	$\bar{X}$	S.D.	t - value
Unprinted GA	0.968	0.074	-0.25
GA + 100% Y - ink	0.974	0.084	
Unprinted MG	0.111	0.020	3.89 <sup>a</sup>
MG + 100% Y - ink	0.089	0.015	

a - The t-value was compared to the values of t-distribution with 38 degrees of freedom, which is approximately  $\pm 2.43$ . This indicates that these two sample macrosmoothness values are significantly different (see Table 4.7 and Figure 4.5).

(b) Microsmoothness (Su).

Sample	Su		
	$\bar{X}$	S.D.	t - value
Unprinted GA	0.951	0.037	1.3
GA + 100% Y - ink	0.932	0.052	
Unprinted MG	0.837	0.079	-1.73
MG + 100% Y - ink	0.874	0.056	

No significant differences were observed.

Table F.2(a) Summary of analysis of variance for macrosmoothness (Sm) of gloss art printed with heatset ink and 'yellow emulsion inks'.

Source of variation	D.F.	Sum of Squares	Mean Squares	F Ratio
Between Groups	3	0.4290	0.1430	17.5581
Within Groups	76	0.6190	0.0081	
Total	79	1.0480		

The F ratio was compared to the value of the F-distribution with 3 and 76 degrees of freedom, which is approximately 2.73. This indicates that the macrosmoothness values between these gloss art prints are significantly different (see Table 4.8 and Figure 4.10).

Table F.2(b) Multiple comparison test - Duncan procedure for Sm of gloss art printed with heatset yellow ink and 'yellow emulsion inks'.

GA/Sm	GA + 90% Y - ink : 10% F.S.	GA + 80% Y - ink : 20% F.S.	GA + 70% Y - ink : 30% F.S.
GA + 100% Y - ink			*
GA + 90% Y - ink : 10% F.S.			*
GA + 80% Y - ink : 20% F.S.			*
GA + 70% Y - ink : 30% F.S.			

\* Denotes pair of samples with a significant difference at the 0.05 level (see Table 4.8 and Figure 4.10).

Table F.3(a) Summary of analysis of variance for microsmoothness (Su) of gloss art printed with heatset ink and 'yellow emulsion inks'.

Source of variation	D.F.	Sum of Squares	Mean Squares	F Ratio
Between Groups	3	0.0066	0.0022	0.9588
Within Groups	76	0.1749	0.0023	
Total	79	0.1815		

The F ratio was compared to the value of the F-distribution with 3 and 76 degrees of freedom, which is approximately 2.73. This indicates that the microsmoothness values between these gloss art prints are not significantly different. Therefore the multiple comparison test - Duncan procedure - was not made (see Table 4.8 and Figure 4.10).

Table F.4(a) Summary of analysis of variance for macrosmoothness (Sm) of machine glazed printed with heatset ink and 'yellow emulsion inks'.

Source of variation	D.F.	Sum of Squares	Mean Squares	F Ratio
Between Groups	3	0.0105	0.0035	16.3850
Within Groups	76	0.0163	0.0002	
Total	79	0.0268		

The F ratio was compared to the value of the F-distribution with 3 and 76 degrees of freedom, which is approximately 2.73. This indicates that the macrosmoothness values between these machine glazed prints are significantly different (see Table 4.9 and Figure 4.11).

Table F.4(b) Multiple comparison test - Duncan procedure for Sm of machine glazed printed with heatset yellow ink and 'yellow emulsion inks'.

MG/Sm	MG + 90% Y - ink : 10% F.S.	MG + 80% Y - ink : 20% F.S.	MG + 70% Y - ink : 30% F.S.
MG + 100% Y - ink		*	*
MG + 90% Y - ink : 10% F.S.		*	*
MG + 80% Y - ink : 20% F.S.			
MG + 70% Y - ink : 30% F.S.			

\* Denotes pair of samples with a significant difference at the 0.05 level (see Table 4.9 and Figure 4.11).

Table F.5(a) Summary of Analysis of variance for microsmoothness (Su) of machine glazed printed with heatset ink and 'yellow emulsion inks'.

Source of variation	D.F.	Sum of Squares	Mean Squares	F Ratio
Between Groups	3	0.0100	0.0033	0.4764
Within Groups	76	0.5291	0.0070	
Total	79	0.5391		

The F ratio was compared to the value of the F-distribution with 3 and 76 degrees of freedom, which is approximately 2.73. This indicates that the microsmoothness values between these machine glazed prints are not significantly different. Therefore, the multiple comparison test - Duncan procedure - was not made (see Table 4.9 and Figure 4.11).



Table F.6(a) Summary of analysis of variance for macrosmoothness (Sm) of gloss art printed with heatset yellow ink, unpigmented ink and their corresponding 'emulsion inks'.

Source of variation	D.F.	Sum of Squares	Mean Squares	F Ratio
Between Groups	3	1.4032	0.4677	42.6033
Within Groups	76	0.8344	0.0110	
Total	79	2.2376		

The F ratio was compared to the value of the F-distribution with 3 and 76 degrees of freedom, which is approximately 2.73. This indicates that the macrosmoothness values between these gloss art prints are significantly different (see Table 4.13 and Figure 4.17).

Table F.6(b) Multiple comparison test - Duncan procedure for Sm of gloss art printed with heatset yellow ink, unpigmented ink and their corresponding 'emulsion inks'.

GA/Sm	GA + 70% Y - ink : 30% F.S.	GA + 100% U - ink	GA + 70% U - ink : 30% F.S.
GA + 100% Y - ink	*	*	*
GA + 70% Y - ink : 30% F.S.			
GA + 100% U - ink	*		
GA + 70% U - ink : 30% F.S.	*		

\* Denotes pair of samples with a significant difference at the 0.05 level (see Table 4.13 and Figure 4.17).

Table F.7(a) Summary of analysis of variance for microsmoothness (Su) of gloss art printed with heatset yellow ink, unpigmented ink and their corresponding 'emulsion inks'.

Source of variation	D.F.	Sum of Squares	Mean Squares	F Ratio
Between Groups	3	0.0136	0.0045	2.3449
Within Groups	76	0.1471	0.0019	
Total	79	0.1607		

The F ratio was compared to the value of the F-distribution with 3 and 76 degrees of freedom, which is approximately 2.73. This indicates that the microsmoothness values between these gloss art prints are not significantly different. Therefore, the multiple comparison test - Duncan procedure - was not made (see Table 4.13 and Figure 4.17).

Table F.8(a) Summary of analysis of variance for macrosmoothness (Sm) of machine glazed printed with heatset yellow ink, unpigmented ink and their corresponding 'emulsion inks'.

Source of variation	D.F.	Sum of Squares	Mean Squares	F Ratio
Between Groups	3	0.0291	0.0097	32.2457
Within Groups	76	0.0228	0.0003	
Total	79	0.0519		

The F ratio was compared to the value of the F-distribution with 3 and 76 degrees of freedom, which is approximately 2.73. This indicates that the macrosmoothness values between these gloss art prints are significantly different (see Table 4.14 and Figure 4.18).

Table F.8(b) Multiple comparison test - Duncan procedure for Sm of machine glazed printed with heatset yellow ink, unpigmented ink and their corresponding 'emulsion inks'.

MG/Sm	MG + 70% Y - ink : 30% F.S.	MG + 100% U - ink	MG + 70% U - ink : 30% F.S.
MG + 100% Y - ink	*	*	*
MG + 70% Y - ink : 30% F.S.			
MG + 100% U - ink	*		*
MG + 70% U - ink : 30% F.S.	*		

\* Denotes pair of samples with a significant difference at the 0.05 level (see Table 4.14 and Figure 4.18).

Table F.9(a) Summary of analysis of variance for microsmoothness (Su) of machine glazed printed with heatset yellow ink, unpigmented ink and their corresponding 'emulsion inks'.

Source of variation	D.F.	Sum of Squares	Mean Squares	F Ratio
Between Groups	3	0.0091	0.0030	0.4865
Within Groups	76	0.4751	0.0063	
Total	79	0.4843		

The F ratio was compared to the value of the F-distribution with 3 and 76 degrees of freedom, which is approximately 2.73. This indicates that the microsmoothness values between these machine glazed prints are not significantly different. Therefore, the multiple comparison test - Duncan procedure - was not made (see Table 4.14 and Figure 4.18).

## Appendix G.

Table G.1 An example, unprinted gloss art; reflectance data and smoothness calculations.

Region	Rp	Rd	Rs	Dp	Ds	Dp×Dp	Ds×Ds	Dp×Ds
14(80)	20	0.001	19.999					
13(75)	9.3	0.001	9.299	2.14	2.62		6.8644	5.6068
12(70)	4.5	0.001	4.499	0.96	1.98		3.9204	1.9008
11(65)	1.8	0.002	1.798	0.5402	1.49		2.2201	0.804898
10(60)	0.9	0.002	0.898	0.18	1.13		1.2769	0.2034
9(55)	0.57	0.002	0.568	0.066	0.86		0.7396	0.05676
8(50)	0.37	0.003	0.367	0.0402	0.65		0.4225	0.02613
7(45)	0.26	0.003	0.257	0.022	0.5		0.25	0.011
6(40)	0.18	0.004	0.176	0.0162	0.38	0.000262	0.1444	0.006156
5(35)	0.12	0.004	0.116	0.012	0.29	0.000144	0.0841	0.00348
4(30)	0.096	0.004	0.092	0.0048	0.23	2.30E-05	0.0529	0.001104
3(25)	0.071	0.004	0.067	0.005	0.17	0.000025	0.0289	0.00085
2(20)	0.056	0.004	0.052	0.003	0.13	0.000009	0.0169	0.00039
1(15)	0.044	0.005	0.039	0.0026	0.09	6.76E-06	0.0081	0.000234

$$C|Dp \times Ds| = \overline{Dp \times Ds} - \overline{Dp} \times \overline{Ds}$$

$$S|Dp| = \sqrt{\overline{Dp^2} - (\overline{Dp})^2}$$

$$S|Ds| = \sqrt{\overline{Ds^2} - (\overline{Ds})^2}$$

$$V|Ds| = \overline{Ds^2} - (\overline{Ds})^2$$

$$S_m = k \frac{C|Dp \times Ds|}{V|Ds|}; \text{ region } \geq 7 = \frac{0.48622}{0.5016} = 0.969338$$

$$S_u = k \frac{C|Dp \times Ds|}{S|Dp| \times S|Ds|}; \text{ region } \leq 6 = \frac{0.000473}{0.005057 \times 0.098} = 0.95518$$

Table G.2 An example of gloss art printed with a heatset yellow ink; reflectance data and smoothness calculations.

Region	Rp	Rd	Rs	Dp	Ds	Dp×Dp	Ds×Ds	Dp×Ds
14(80)	21	0.002	20.998					
13(75)	10	0.002	9.998	2.2	2.62		6.8644	5.764
12(70)	5.8	0.002	5.798	0.84	1.98		3.9204	1.6632
11(65)	2.6	0.003	2.597	0.6402	1.49		2.2201	0.953898
10(60)	1.6	0.003	1.597	0.2	1.13		1.2769	0.226
9(55)	1.1	0.003	1.097	0.1	0.86		0.7396	0.086
8(50)	0.8	0.003	0.797	0.06	0.65		0.4225	0.039
7(45)	0.54	0.004	0.536	0.0522	0.5		0.25	0.0261
6(40)	0.36	0.004	0.356	0.036	0.38	0.001296	0.1444	0.01368
5(35)	0.23	0.004	0.226	0.026	0.29	0.000676	0.0841	0.00754
4(30)	0.18	0.004	0.176	0.01	0.23	0.0001	0.0529	0.0023
3(25)	0.16	0.005	0.155	0.0042	0.17	1.764E-05	0.0289	0.000714
2(20)	0.14	0.005	0.135	0.004	0.13	0.000016	0.0169	0.00052
1(15)	0.1	0.005	0.095	0.008	0.09	0.000064	0.0081	0.00072

$$C|Dp \times Ds| = \overline{Dp \times Ds} - \overline{Dp} \overline{Ds}$$

$$S|Dp| = \sqrt{\overline{Dp^2} - (\overline{Dp})^2}$$

$$S|Ds| = \sqrt{\overline{Ds^2} - (\overline{Ds})^2}$$

$$V|Ds| = \overline{Ds^2} - (\overline{Ds})^2$$

$$S_m = k \frac{C|Dp \times Ds|}{V|Ds|}; \text{ region } \geq 7 = \frac{0.480297}{0.5016} = 0.957529$$

$$S_u = k \frac{C|Dp \times Ds|}{S|Dp| \times S|Ds|}; \text{ region } \leq 6 = \frac{0.001085}{0.012063 \times 0.098} = 0.917939$$

Table G.3 An example of gloss art printed with 10% water content 'yellow emulsion ink'; reflectance data and smoothness calculations.

Region	Rp	Rd	Rs	Dp	Ds	Dp×Dp	Ds×Ds	Dp×Ds
14(80)	26	0.002	25.998					
13(75)	14	0.002	13.998	2.4	2.62		6.8644	6.288
12(70)	7.4	0.002	7.398	1.32	1.98		3.9204	2.6136
11(65)	4.9	0.002	4.898	0.5	1.49		2.2201	0.745
10(60)	2.8	0.003	2.797	0.4202	1.13		1.2769	0.474826
9(55)	1.5	0.003	1.497	0.26	0.86		0.7396	0.2236
8(50)	0.92	0.003	0.917	0.116	0.65		0.4225	0.0754
7(45)	0.62	0.003	0.617	0.06	0.5		0.25	0.03
6(40)	0.39	0.004	0.386	0.0462	0.38	0.0021344	0.1444	0.017556
5(35)	0.31	0.004	0.306	0.016	0.29	0.000256	0.0841	0.00464
4(30)	0.25	0.004	0.246	0.012	0.23	0.000144	0.0529	0.00276
3(25)	0.22	0.004	0.216	0.006	0.17	0.000036	0.0289	0.00102
2(20)	0.25	0.004	0.246	-0.006	0.13	0.000036	0.0169	-0.00078
1(15)	0.27	0.004	0.266	-0.004	0.09	0.000016	0.0081	-0.00036

$$C|Dp \times Ds| = \overline{Dp \times Ds} - \overline{Dp} \times \overline{Ds}$$

$$S|Dp| = \sqrt{Dp^2 - (\overline{Dp})^2}$$

$$S|Ds| = \sqrt{Ds^2 - (\overline{Ds})^2}$$

$$V|Ds| = \overline{Ds^2} - (\overline{Ds})^2$$

$$S_m = k \frac{C|Dp \times Ds|}{V|Ds|}; \text{ region } \geq 7 = \frac{0.536728}{0.5016} = 1.070031$$

$$S_u = k \frac{C|Dp \times Ds|}{S|Dp| \times S|Ds|}; \text{ region } \leq 6 = \frac{0.001624}{0.017326 \times 0.098} = 0.956362$$

Table G.4 An example of gloss art printed with 20% water content 'yellow emulsion ink'; reflectance data and smoothness calculations.

Region	Rp	Rd	Rs	Dp	Ds	Dp×Dp	Ds×Ds	Dp×Ds
14(80)	23	0.002	22.998					
13(75)	11	0.001	10.999	2.3998	2.62		6.8644	6.287476
12(70)	6.6	0.002	6.598	0.8802	1.98		3.9204	1.742796
11(65)	3.7	0.002	3.698	0.58	1.49		2.2201	0.8642
10(60)	2.1	0.003	2.097	0.3202	1.13		1.2769	0.361826
9(55)	1.2	0.003	1.197	0.18	0.86		0.7396	0.1548
8(50)	0.86	0.003	0.857	0.068	0.65		0.4225	0.0442
7(45)	0.65	0.003	0.647	0.042	0.5		0.25	0.021
6(40)	0.5	0.003	0.497	0.03	0.38	0.0009	0.1444	0.0114
5(35)	0.43	0.003	0.427	0.014	0.29	0.000196	0.0841	0.00406
4(30)	0.37	0.004	0.366	0.0122	0.23	0.000149	0.0529	0.002806
3(25)	0.32	0.004	0.316	0.01	0.17	0.0001	0.0289	0.0017
2(20)	0.29	0.004	0.286	0.006	0.13	0.000036	0.0169	0.00078
1(15)	0.26	0.004	0.256	0.006	0.09	0.000036	0.0081	0.00054

$$C|Dp \times Ds| = \overline{Dp \times Ds} - \overline{Dp} \times \overline{Ds}$$

$$S|Dp| = \sqrt{\overline{Dp^2} - (\overline{Dp})^2}$$

$$S|Ds| = \sqrt{\overline{Ds^2} - (\overline{Ds})^2}$$

$$V|Ds| = \overline{Ds^2} - (\overline{Ds})^2$$

$$S_m = k \frac{C|Dp \times Ds|}{V|Ds|}; \text{ region } \geq 7 = \frac{0.511717}{0.5016} = 1.02017$$

$$S_u = k \frac{C|Dp \times Ds|}{S|Dp| \times S|Ds|}; \text{ region } \leq 6 = \frac{0.000746}{0.008141 \times 0.098} = 0.934449$$



Table G.5 An example of gloss art printed with 30% water content 'yellow emulsion ink'; reflectance data and smoothness calculations.

Region	Rp	Rd	Rs	Dp	Ds	Dp×Dp	Ds×Ds	Dp×Ds
14(80)	21	0.001	20.999					
13(75)	10	0.001	9.999	2.2	2.62		6.8644	5.764
12(70)	7.4	0.002	7.398	0.5202	1.98		3.9204	1.029996
11(65)	5	0.002	4.998	0.48	1.49		2.2201	0.7152
10(60)	3	0.003	2.997	0.4002	1.13		1.2769	0.452226
9(55)	2.1	0.003	2.097	0.18	0.86		0.7396	0.1548
8(50)	1.6	0.004	1.596	0.1002	0.65		0.4225	0.06513
7(45)	1.1	0.004	1.096	0.1	0.5		0.25	0.05
6(40)	0.83	0.004	0.826	0.054	0.38	0.002916	0.1444	0.02052
5(35)	0.53	0.004	0.526	0.06	0.29	0.0036	0.0841	0.0174
4(30)	0.38	0.004	0.376	0.03	0.23	0.0009	0.0529	0.0069
3(25)	0.26	0.005	0.255	0.0242	0.17	0.000586	0.0289	0.004114
2(20)	0.21	0.005	0.205	0.01	0.13	0.0001	0.0169	0.0013
1(15)	0.2	0.005	0.195	0.002	0.09	0.000004	0.0081	0.00018

$$C | Dp \times Ds | = \overline{Dp \times Ds} - \overline{Dp} \times \overline{Ds}$$

$$S | Dp | = \sqrt{\overline{Dp^2} - (\overline{Dp})^2}$$

$$S | Ds | = \sqrt{\overline{Ds^2} - (\overline{Ds})^2}$$

$$V | Ds | = \overline{Ds^2} - (\overline{Ds})^2$$

$$S_m = k \frac{C | Dp \times Ds |}{V | Ds |} ; \text{region} \geq 7 = \frac{0.426092}{0.5016} = 0.849466$$

$$S_u = k \frac{C | Dp \times Ds |}{S | Dp | \times S | Ds |} ; \text{region} \leq 6 = \frac{0.001945}{0.021188 \times 0.098} = 0.936779$$

Table G.6 An example of gloss art printed with an unpigmented ink; reflectance data and smoothness calculations.

Region	Rp	Rd	Rs	Dp	Ds	Dp×Dp	Ds×Ds	Dp×Ds
14(80)	29	0.001	28.999					
13(75)	16	0.001	15.999	2.6	2.62		6.8644	6.812
12(70)	11	0.002	10.998	1.0002	1.98		3.9204	1.980396
11(65)	4.1	0.002	4.098	1.38	1.49		2.2201	2.0562
10(60)	3.2	0.002	3.198	0.18	1.13		1.2769	0.2034
9(55)	2.2	0.003	2.197	0.2002	0.86		0.7396	0.172172
8(50)	1.7	0.003	1.697	0.1	0.65		0.4225	0.065
7(45)	1.3	0.003	1.297	0.08	0.5		0.25	0.04
6(40)	1	0.004	0.996	0.0602	0.38	0.003624	0.1444	0.022876
5(35)	0.78	0.004	0.776	0.044	0.29	0.001936	0.0841	0.01276
4(30)	0.59	0.004	0.586	0.038	0.23	0.001444	0.0529	0.00874
3(25)	0.39	0.004	0.386	0.04	0.17	0.0016	0.0289	0.0068
2(20)	0.27	0.005	0.265	0.0242	0.13	0.0005856	0.0169	0.003146
1(15)	0.2	0.005	0.195	0.014	0.09	0.000196	0.0081	0.00126

$$C|Dp \times Ds| = \overline{Dp \times Ds} - \overline{Dp} \times \overline{Ds}$$

$$S|Dp| = \sqrt{Dp^2 - (\overline{Dp})^2}$$

$$S|Ds| = \sqrt{Ds^2 - (\overline{Ds})^2}$$

$$V|Ds| = \overline{Ds^2} - (\overline{Ds})^2$$

$$S_m = k \frac{C|Dp \times Ds|}{V|Ds|}; \text{ region } \geq 7 = \frac{0.574822}{0.5016} = 1.145977$$

$$S_u = k \frac{C|Dp \times Ds|}{S|Dp| \times S|Ds|}; \text{ region } \leq 6 = \frac{0.001366}{0.014661 \times 0.098} = 0.950744$$

Table G.7 An example of gloss art printed with 30% water content 'unpigmented emulsion ink'; reflectance data and smoothness calculations.

Region	Rp	Rd	Rs	Dp	Ds	Dp×Dp	Ds×Ds	Dp×Ds
14(80)	30	0.002	29.998					
13(75)	14	0.003	13.997	3.2002	2.62		6.8644	8.384524
12(70)	11	0.003	10.997	0.6	1.98		3.9204	1.188
11(65)	7.9	0.003	7.897	0.62	1.49		2.2201	0.9238
10(60)	4.4	0.004	4.396	0.7002	1.13		1.2769	0.791226
9(55)	2.7	0.004	2.696	0.34	0.86		0.7396	0.2924
8(50)	1.6	0.004	1.596	0.22	0.65		0.4225	0.143
7(45)	1.3	0.004	1.296	0.06	0.5		0.25	0.03
6(40)	0.92	0.005	0.915	0.0762	0.38	0.0058064	0.1444	0.028956
5(35)	0.56	0.005	0.555	0.072	0.29	0.005184	0.0841	0.02088
4(30)	0.42	0.005	0.415	0.028	0.23	0.000784	0.0529	0.00644
3(25)	0.41	0.006	0.404	0.0022	0.17	4.84E-06	0.0289	0.000374
2(20)	0.36	0.006	0.354	0.01	0.13	0.0001	0.0169	0.0013
1(15)	0.33	0.006	0.324	0.006	0.09	0.000036	0.0081	0.00054

$$C|Dp \times Ds| = \overline{Dp \times Ds} - \overline{Dp} \times \overline{Ds}$$

$$S|Dp| = \sqrt{\overline{Dp^2} - (\overline{Dp})^2}$$

$$S|Ds| = \sqrt{\overline{Ds^2} - (\overline{Ds})^2}$$

$$V|Ds| = \overline{Ds^2} - (\overline{Ds})^2$$

$$S_m = k \frac{C|Dp \times Ds|}{V|Ds|} ; \text{region} \geq 7 = \frac{0.597689}{0.5016} = 1.191565$$

$$S_u = k \frac{C|Dp \times Ds|}{S|Dp| \times S|Ds|} ; \text{region} \leq 6 = \frac{0.002782}{0.030596 \times 0.098} = 0.927934$$

Table G.8 An example, unprinted machine glazed; reflectance data and smoothness calculations.

Region	Rp	Rd	Rs	Dp	Ds	Dp×Dp	Ds×Ds	Dp×Ds
14(80)	2.3	0.002	2.298					
13(75)	1	0.003	0.997	0.2602	2.62		6.8644	0.681724
12(70)	0.5	0.003	0.497	0.1	1.98		3.9204	0.198
11(65)	0.27	0.003	0.267	0.046	1.49		2.2201	0.06854
10(60)	0.18	0.003	0.177	0.018	1.13		1.2769	0.02034
9(55)	0.11	0.004	0.106	0.0142	0.86		0.7396	0.012212
8(50)	0.068	0.005	0.063	0.0086	0.65		0.4225	0.00559
7(45)	0.05	0.005	0.045	0.0036	0.5		0.25	0.0018
6(40)	0.04	0.006	0.034	0.0022	0.38	4.84E-06	0.1444	0.000836
5(35)	0.036	0.005	0.031	0.0006	0.29	3.6E-07	0.0841	0.000174
4(30)	0.029	0.004	0.025	0.0012	0.23	1.44E-06	0.0529	0.000276
3(25)	0.026	0.004	0.022	0.0006	0.17	3.6E-07	0.0289	0.000102
2(20)	0.024	0.004	0.02	0.0004	0.13	1.6E-07	0.0169	0.000052
1(15)	0.032	0.004	0.028	-0.0016	0.09	2.56E-06	0.0081	-0.000144

$$C|Dp \times Ds| = \overline{Dp \times Ds} - \overline{Dp} \times \overline{Ds}$$

$$S|Dp| = \sqrt{\overline{Dp^2} - (\overline{Dp})^2}$$

$$S|Ds| = \sqrt{\overline{Ds^2} - (\overline{Ds})^2}$$

$$V|Ds| = \overline{Ds^2} - (\overline{Ds})^2$$

$$S_m = k \frac{C|Dp \times Ds|}{V|Ds|}; \text{ region } \geq 7 = \frac{0.056294}{0.5016} = 0.112229$$

$$S_u = k \frac{C|Dp \times Ds|}{S|Dp| \times S|Ds|}; \text{ region } \leq 6 = \frac{0.0000942}{0.00114 \times 0.098} = 0.843112$$

Table G.9 An example, machine glazed printed with a heatset yellow ink; reflectance data and smoothness calculations.

Region	Rp	Rd	Rs	Dp	Ds	Dp×Dp	Ds×Ds	Dp×Ds
14(80)	2.3	0.002	2.298					
13(75)	1.3	0.003	1.297	0.2002	2.62		6.8644	0.524524
12(70)	0.86	0.003	0.857	0.088	1.98		3.9204	0.17424
11(65)	0.29	0.003	0.287	0.114	1.49		2.2201	0.16986
10(60)	0.22	0.004	0.216	0.0142	1.13		1.2769	0.016046
9(55)	0.17	0.004	0.166	0.01	0.86		0.7396	0.0086
8(50)	0.094	0.004	0.09	0.0152	0.65		0.4225	0.00988
7(45)	0.064	0.004	0.06	0.006	0.5		0.25	0.003
6(40)	0.034	0.005	0.029	0.0062	0.38	3.844E-05	0.1444	0.002356
5(35)	0.025	0.005	0.02	0.0018	0.29	3.24E-06	0.0841	0.000522
4(30)	0.024	0.006	0.018	0.0004	0.23	1.6E-07	0.0529	0.000092
3(25)	0.016	0.005	0.011	0.0014	0.17	1.96E-06	0.0289	0.000238
2(20)	0.016	0.006	0.01	0.0002	0.13	4E-08	0.0169	0.000026
1(15)	0.022	0.006	0.016	-0.0012	0.09	1.44E-06	0.0081	-0.000108

$$C|Dp \times Ds| = \overline{Dp \times Ds} - \overline{Dp} \times \overline{Ds}$$

$$S|Dp| = \sqrt{Dp^2 - (\overline{Dp})^2}$$

$$S|Ds| = \sqrt{Ds^2 - (\overline{Ds})^2}$$

$$V|Ds| = \overline{Ds^2} - (\overline{Ds})^2$$

$$S_m = k \frac{C|Dp \times Ds|}{V|Ds|}; \text{ region } \geq 7 = \frac{0.045137}{0.5016} = 0.089986$$

$$S_u = k \frac{C|Dp \times Ds|}{S|Dp| \times S|Ds|}; \text{ region } \leq 6 = \frac{0.000206}{0.002323 \times 0.098} = 0.903483$$

Table G.10 An example, machine glazed printed with 10% water content 'yellow enulsion ink'; reflectance data and smoothness calculations.

Region	Rp	Rd	Rs	Dp	Ds	Dp×Dp	Ds×Ds	Dp×Ds
14(80)	1.6	0.001	1.599					
13(75)	0.53	0.001	0.529	0.214	2.62		6.8644	0.56068
12(70)	0.23	0.002	0.228	0.0602	1.98		3.9204	0.119196
11(65)	0.11	0.002	0.108	0.024	1.49		2.2201	0.03576
10(60)	0.06	0.003	0.057	0.0102	1.13		1.2769	0.011526
9(55)	0.042	0.003	0.039	0.0036	0.86		0.7396	0.003096
8(50)	0.04	0.003	0.037	0.0004	0.65		0.4225	0.00026
7(45)	0.032	0.003	0.029	0.0016	0.5		0.25	0.0008
6(40)	0.025	0.004	0.021	0.0016	0.38	2.56E-06	0.1444	0.000608
5(35)	0.015	0.004	0.011	0.002	0.29	0.000004	0.0841	0.00058
4(30)	0.011	0.004	0.007	0.0008	0.23	6.4E-07	0.0529	0.000184
3(25)	0.009	0.004	0.005	0.0004	0.17	1.6E-07	0.0289	0.000068
2(20)	0.01	0.005	0.005	-1.73E-19	0.13	3.01E-38	0.0169	-2.26E-20
1(15)	0.009	0.004	0.005	1.73E-19	0.09	3.01E-38	0.0081	1.56E-20

$$C|Dp \times Ds| = \overline{Dp \times Ds} - \overline{Dp} \times \overline{Ds}$$

$$S|Dp| = \sqrt{\overline{Dp^2} - (\overline{Dp})^2}$$

$$S|Ds| = \sqrt{\overline{Ds^2} - (\overline{Ds})^2}$$

$$V|Ds| = \overline{Ds^2} - (\overline{Ds})^2$$

$$S_m = k \frac{C|Dp \times Ds|}{V|Ds|}; \text{ region } \geq 7 = \frac{0.045327}{0.5016} = 0.090364$$

$$S_u = k \frac{C|Dp \times Ds|}{S|Dp| \times S|Ds|}; \text{ region } \leq 6 = \frac{0.000068}{0.000766 \times 0.098} = 0.905914$$

Table G.11 An example, machine glazed printed with 20% water content 'yellow enulsion ink'; reflectance data and smoothness calculations.

Region	Rp	Rd	Rs	Dp	Ds	Dp×Dp	Ds×Ds	Dp×Ds
14(80)	1.3	0.001	1.299					
13(75)	0.52	0.002	0.518	0.1562	2.62		6.8644	0.409244
12(70)	0.29	0.002	0.288	0.046	1.98		3.9204	0.09108
11(65)	0.14	0.002	0.138	0.03	1.49		2.2201	0.0447
10(60)	0.084	0.003	0.081	0.0114	1.13		1.2769	0.012882
9(55)	0.076	0.003	0.073	0.0016	0.86		0.7396	0.001376
8(50)	0.063	0.003	0.06	0.0026	0.65		0.4225	0.00169
7(45)	0.045	0.003	0.042	0.0036	0.5		0.25	0.0018
6(40)	0.028	0.004	0.024	0.0036	0.38	1.296E-05	0.1444	0.001368
5(35)	0.018	0.004	0.014	0.002	0.29	0.000004	0.0841	0.00058
4(30)	0.016	0.004	0.012	0.0004	0.23	1.6E-07	0.0529	0.000092
3(25)	0.015	0.004	0.011	0.0002	0.17	4E-08	0.0289	0.000034
2(20)	0.015	0.004	0.011	0	0.13	0	0.0169	0
1(15)	0.01	0.004	0.006	0.001	0.09	0.000001	0.0081	0.00009

$$C|Dp \times Ds| = \overline{Dp \times Ds} - \overline{Dp} \times \overline{Ds}$$

$$S|Dp| = \sqrt{\overline{Dp^2} - (\overline{Dp})^2}$$

$$S|Ds| = \sqrt{\overline{Ds^2} - (\overline{Ds})^2}$$

$$V|Ds| = \overline{Ds^2} - (\overline{Ds})^2$$

$$S_m = k \frac{C|Dp \times Ds|}{V|Ds|}; \text{ region } \geq 7 = \frac{0.03304}{0.5016} = 0.06587$$

$$S_u = k \frac{C|Dp \times Ds|}{S|Dp| \times S|Ds|}; \text{ region } \leq 6 = \frac{0.000103}{0.00126 \times 0.098} = 0.831688$$

Table G.12 An example, machine glazed printed with 30% water content 'yellow enulsion ink'; reflectance data and smoothness calculations.

Region	Rp	Rd	Rs	Dp	Ds	Dp×Dp	Ds×Ds	Dp×Ds
14(80)	1.3	0.002	1.298					
13(75)	0.51	0.002	0.508	0.158	2.62		6.8644	0.41396
12(70)	0.25	0.003	0.247	0.0522	1.98		3.9204	0.103356
11(65)	0.2	0.003	0.197	0.01	1.49		2.2201	0.0149
10(60)	0.16	0.003	0.157	0.008	1.13		1.2769	0.00904
9(55)	0.1	0.003	0.097	0.012	0.86		0.7396	0.01032
8(50)	0.06	0.004	0.056	0.0082	0.65		0.4225	0.00533
7(45)	0.035	0.003	0.032	0.0048	0.5		0.25	0.0024
6(40)	0.025	0.004	0.021	0.0022	0.38	4.84E-06	0.1444	0.000836
5(35)	0.018	0.004	0.014	0.0014	0.29	1.96E-06	0.0841	0.000406
4(30)	0.016	0.004	0.012	0.0004	0.23	1.6E-07	0.0529	0.000092
3(25)	0.015	0.004	0.011	0.0002	0.17	4E-08	0.0289	0.000034
2(20)	0.012	0.004	0.008	0.0006	0.13	3.6E-07	0.0169	0.000078
1(15)	0.01	0.004	0.006	0.0004	0.09	1.6E-07	0.0081	0.000036

$$C|Dp \times Ds| = \overline{Dp \times Ds} - \overline{Dp} \times \overline{Ds}$$

$$S|Dp| = \sqrt{\overline{Dp^2} - (\overline{Dp})^2}$$

$$S|Ds| = \sqrt{\overline{Ds^2} - (\overline{Ds})^2}$$

$$V|Ds| = \overline{Ds^2} - (\overline{Ds})^2$$

$$S_m = k \frac{C|Dp \times Ds|}{V|Ds|}; \text{ region } \geq 7 = \frac{0.032206}{0.5016} = 0.064207$$

$$S_u = k \frac{C|Dp \times Ds|}{S|Dp| \times S|Ds|}; \text{ region } \leq 6 = \frac{0.0000607}{0.000709 \times 0.098} = 0.873527$$



Table G.13 An example, machine glazed printed with an unpigmented ink; reflectance data and smoothness calculations.

Region	Rp	Rd	Rs	Dp	Ds	Dp×Dp	Ds×Ds	Dp×Ds
14(80)	2.3	0.001	2.299					
13(75)	1.2	0.002	1.198	0.2202	2.62		6.8644	0.576924
12(70)	0.62	0.002	0.618	0.116	1.98		3.9204	0.22968
11(65)	0.4	0.003	0.397	0.0442	1.49		2.2201	0.065858
10(60)	0.24	0.003	0.237	0.032	1.13		1.2769	0.03616
9(55)	0.16	0.003	0.157	0.016	0.86		0.7396	0.01376
8(50)	0.14	0.003	0.137	0.004	0.65		0.4225	0.0026
7(45)	0.14	0.004	0.136	0.0002	0.5		0.25	0.0001
6(40)	0.1	0.004	0.096	0.008	0.38	0.000064	0.1444	0.00304
5(35)	0.044	0.004	0.04	0.0112	0.29	0.0001254	0.0841	0.003248
4(30)	0.026	0.005	0.021	0.0038	0.23	1.444E-05	0.0529	0.000874
3(25)	0.019	0.005	0.014	0.0014	0.17	1.96E-06	0.0289	0.000238
2(20)	0.015	0.004	0.011	0.0006	0.13	3.6E-07	0.0169	0.000078
1(15)	0.013	0.004	0.009	0.0004	0.09	1.6E-07	0.0081	0.000036

$$C|Dp \times Ds| = \overline{Dp \times Ds} - \overline{Dp} \times \overline{Ds}$$

$$S|Dp| = \sqrt{\overline{Dp^2} - (\overline{Dp})^2}$$

$$S|Ds| = \sqrt{\overline{Ds^2} - (\overline{Ds})^2}$$

$$V|Ds| = \overline{Ds^2} - (\overline{Ds})^2$$

$$Sm = k \frac{C|Dp \times Ds|}{V|Ds|}; \text{ region } \geq 7 = \frac{0.050667}{0.5016} = 0.10101$$

$$Su = k \frac{C|Dp \times Ds|}{S|Dp| \times S|Ds|}; \text{ region } \leq 6 = \frac{0.000342}{0.004059 \times 0.098} = 0.860271$$

Table G.14 An example, machine glazed printed with 30% water content 'unpigmented enulsion ink'; reflectance data and smoothness calculations.

Region	Rp	Rd	Rs	Dp	Ds	Dp×Dp	Ds×Ds	Dp×Ds
14(80)	2.3	0.002	2.298					
13(75)	0.7	0.002	0.698	0.32	2.62		6.8644	0.8384
12(70)	0.51	0.003	0.507	0.0382	1.98		3.9204	0.075636
11(65)	0.26	0.003	0.257	0.05	1.49		2.2201	0.0745
10(60)	0.19	0.003	0.187	0.014	1.13		1.2769	0.01582
9(55)	0.12	0.003	0.117	0.014	0.86		0.7396	0.01204
8(50)	0.1	0.004	0.096	0.0042	0.65		0.4225	0.00273
7(45)	0.045	0.003	0.042	0.0108	0.5		0.25	0.0054
6(40)	0.023	0.004	0.019	0.0046	0.38	2.116E-05	0.1444	0.001748
5(35)	0.021	0.004	0.017	0.0004	0.29	1.6E-07	0.0841	0.000116
4(30)	0.018	0.004	0.014	0.0006	0.23	3.6E-07	0.0529	0.000138
3(25)	0.018	0.004	0.014	0	0.17	0	0.0289	0
2(20)	0.022	0.005	0.017	-0.0006	0.13	3.6E-07	0.0169	-0.000078
1(15)	0.022	0.004	0.018	-0.0002	0.09	4E-08	0.0081	-0.000018

$$C|Dp \times Ds| = \overline{Dp \times Ds} - \overline{Dp} \times \overline{Ds}$$

$$S|Dp| = \sqrt{\overline{Dp^2} - (\overline{Dp})^2}$$

$$S|Ds| = \sqrt{\overline{Ds^2} - (\overline{Ds})^2}$$

$$V|Ds| = \overline{Ds^2} - (\overline{Ds})^2$$

$$S_m = k \frac{C|Dp \times Ds|}{V|Ds|}; \text{ region } \geq 7 = \frac{0.06137}{0.5016} = 0.12234$$

$$S_u = k \frac{C|Dp \times Ds|}{S|Dp| \times S|Ds|}; \text{ region } \leq 6 = \frac{0.000146}{0.001744 \times 0.098} = 0.852506$$